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A SPIN ONE EXCHANGE AND A DYNAMICAL
ISING MODEL OF CRITICAL PHENOMENA

BY

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The undersigned certify that they have read, and recommend to the Faculty of Graduate Studies for acceptance, a thesis entitled A SPIN ONE EXCHANGE AND A DYNAMICAL ISING MODEL OF CRITICAL PHENOMENA, submitted by Grant Alexander Thomas Allan, in partial fulfilment of the requirements for the degree of Doctor of Philosophy.

ABSTRACT

In part I, Schrodinger's spin one exchange interaction, $P_{ij} = (\vec{S}_i \cdot \vec{S}_j)^2 + (\vec{S}_i \cdot \vec{S}_j) - 1$, is introduced to form a Hamiltonian for a nearest neighbour lattice model of ferromagnetism. The physical relevance of the Hamiltonian is discussed. Group theoretic methods together with the cluster expansion technique for obtaining the high temperature partition function and initial susceptibility series are introduced. The first eight terms of the susceptibility series are obtained for the face-centred cubic lattice. Analysis by the ratio and Pade approximant methods yields the critical temperature and the critical index γ which is found to be very nearly $5/4$ and thus certainly different from the spin one Heisenberg model index.

In part II, the Kubo linear response theory is applied to the quantum mechanical Ising model to obtain a general expression which is valid for all regular lattices for the perpendicular response function in terms of spin correlations. For the honeycomb and plane square lattices, exact expressions are found for the frequency dependent initial perpendicular susceptibility $\chi_{\perp}(\omega, T)$ valid for all temperatures. A discrepancy arises between the zero frequency result for the plane square lattice and Fisher's static calculation. This is resolved by using an extended calculation which explicitly illustrates the difference between isothermal and adiabatic susceptibilities.



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Finally the magnetic inelastic scattering of neutrons from a spin $\frac{1}{2}$ Ising system is considered. For the plane square lattice, the exact energy distribution of the scattering for all temperatures is obtained and found to vary most markedly in the critical region. Some qualitative projections are also made concerning three dimensional systems.

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CHAPTER I

INTRODUCTIONA. Survey of Physical Systems

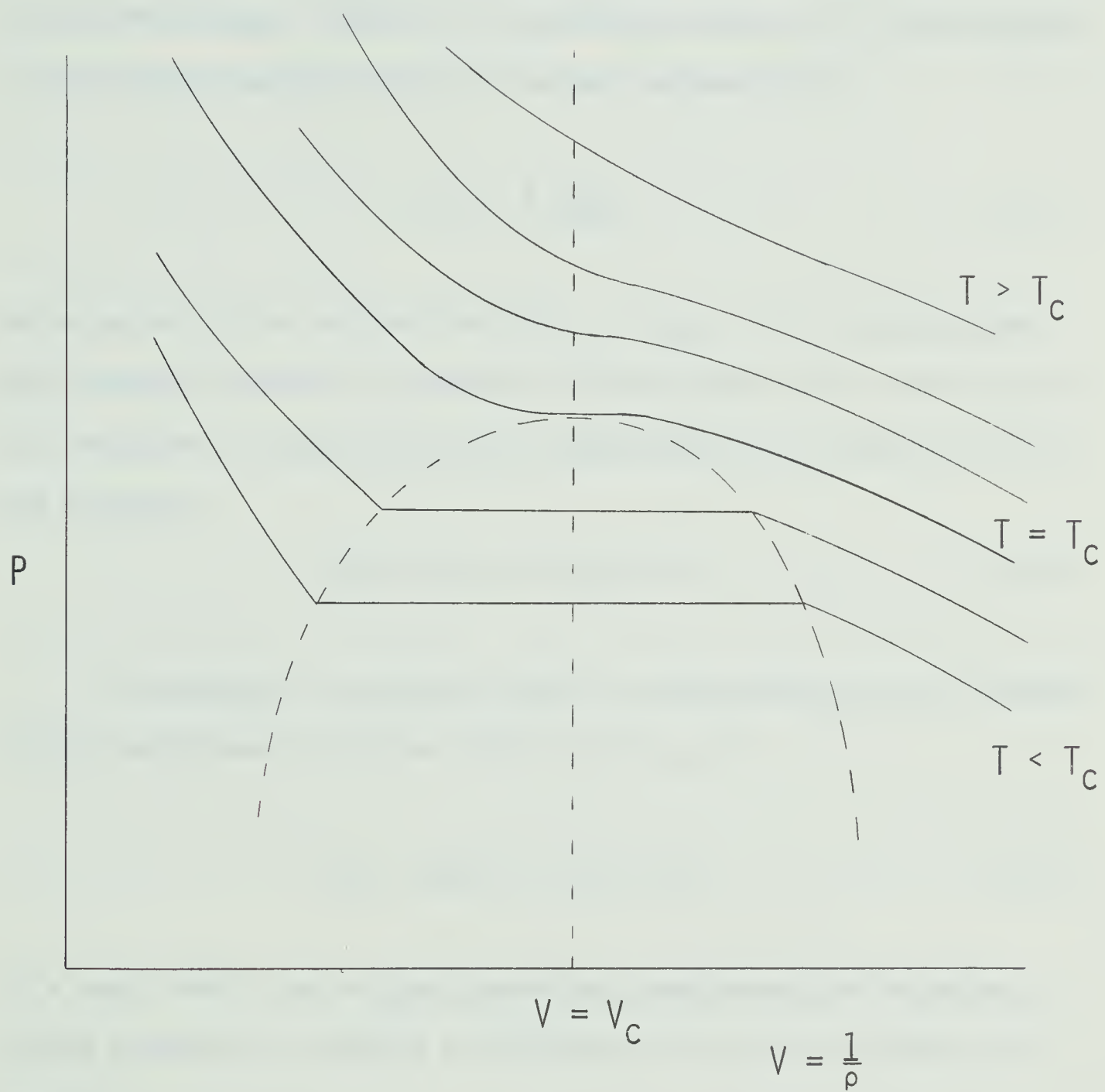
The study of phase transitions and critical points cuts across many of the familiar boundaries of the sciences. It attracts the interest of the solid-state physicist, the low-temperature physicist, the physical chemist, the chemical engineer and the metallurgist. One reason lies in the large variety of physical systems that exhibit critical phenomena. They include simple fluids, ferromagnets, binary solutions, antiferromagnets and binary alloys. It is one of the purposes of these opening paragraphs to point out that despite the variety of systems, their quantitative behavior displays remarkable similarities. It is one of the challenges of this subject to reconcile the variety with the similarity and, as we proceed, we shall see to what extent this has been achieved.

It should be pointed out that such systems as ferroelectrics, superconductors and superfluids have been deliberately left out. They stand apart in the first case because of the dominant nature of the long range coulomb forces and in the latter because of the essentially quantum-mechanical origin of the phenomena.

In figure 1.1 a number of (p,v) isotherms for a simple fluid are

Fig. 1.1

Typical isotherms for a simple fluid.



sketched. It is an experimental fact that for temperatures greater than the critical temperature, T_c , one can pass by compression from the gaseous state to the liquid state without any anomalies in the derivatives of the free energy. However for temperatures close to T_c , there exists a well-defined maximum in the isothermal compressibility,

$$K_T = - \frac{1}{V} \left(\frac{\partial V}{\partial p} \right)_T , \quad (1.1)$$

which occurs at the critical density $\rho_c = 1/v_c$. As T_c is approached, this maximum increases in intensity (corresponding to the flattening of the isotherms in figure 1.1) and we characterize the critical point by the statement,

$$K_T(\rho = \rho_c, T \rightarrow T_c+) \rightarrow \infty . \quad (1.2)$$

In addition, the critical point is characterized by the 'closure' of the coexistence curve (see figure 1.1) i.e. by

$$(\rho_L - \rho_G) \rightarrow 0 . \quad (T \rightarrow T_c-) \quad (1.3)$$

In a very careful and accurate experiment, Weinberger and Schneider (1952) measured this density discontinuity for Xenon and obtained the result,

$$(\rho_L - \rho_G) = A(T_c - T)^{\beta} , \quad (T \rightarrow T_c-) \quad (1.4)$$

with $\beta = 0.345 \pm 0.015$. (1.5)

There is an understandable tendency in this field to anticipate that the critical indices, of which ' β ' is our first example, should equal rational numbers. In that spirit (1.5) strongly suggests that a ' $1/3$ ' rd power law describes the behavior of the coexistence curve.

Finally, we add that the critical point is also associated with a striking thermal anomaly. In recent experiments on the specific heat of argon, Bagatskii, Voronel and Gusak (1963) reach the conclusion that

$$C_v(T) \rightarrow \infty. (T \rightarrow T_c \pm, \rho = \rho_c) \quad (1.6)$$

The above experimental facts indicate the critical region is of primary importance and interest. This is more than substantiated when we consider ferromagnets which would appear to bear very little similarity in their physical properties to simple fluids. A ferromagnet is characterized by a spontaneous or residual magnetization which persists at low temperatures even when the magnetizing field is reduced to zero, i.e.

$$M_0(T) = \lim_{H \rightarrow 0+} M(T, H) \quad (1.7)$$

where $M(T, H)$ is the equilibrium magnetization in the field H .

The critical temperature (known as the Curie point in this context)

is associated with the following behavior, (see figure 1.2)

$$\begin{aligned} M_0(T) &\longrightarrow 0 & (T \longrightarrow T_c^-) \\ M_0(T) &= 0 & T \geq T_c \end{aligned} \quad (1.8)$$

In addition, the initial susceptibility, given by

$$\chi_0(T) = \left(\frac{\partial M}{\partial H} \right)_{T, H=0}, \quad (1.9)$$

is positive and as $T \longrightarrow T_c^+$, it diverges to infinity (see figure 1.2). Reflection of the magnetization curve in figure 1.2 about the $M_0 = 0$ axis and anticlockwise rotation through an angle of 90° suggests an analogy with the gas-liquid coexistence curve in figure 1.1. In figure 1.3 an ideal magnetization curve for a ferromagnet is drawn and again a comparison with figure 1.1 suggests that the susceptibility of the ferromagnet and the compressibility of the gas-liquid system are analogous variables. One difference, however, is that the discontinuity in the magnetization, i.e. a two phase region, occurs only for zero field while $\rho_L - \rho_G$ is non-zero for all $p < p_c$. (In both instances, of course, T must be less than T_c).

That the above comparison is somewhat more than an analogy is demonstrated by the experiments of Heller and Benedek (1965). Using magnetic resonance techniques, they measured the spontaneous magnetization

Fig. 1.2

Spontaneous magnetization and initial susceptibility
of a ferromagnet near its Curie point.

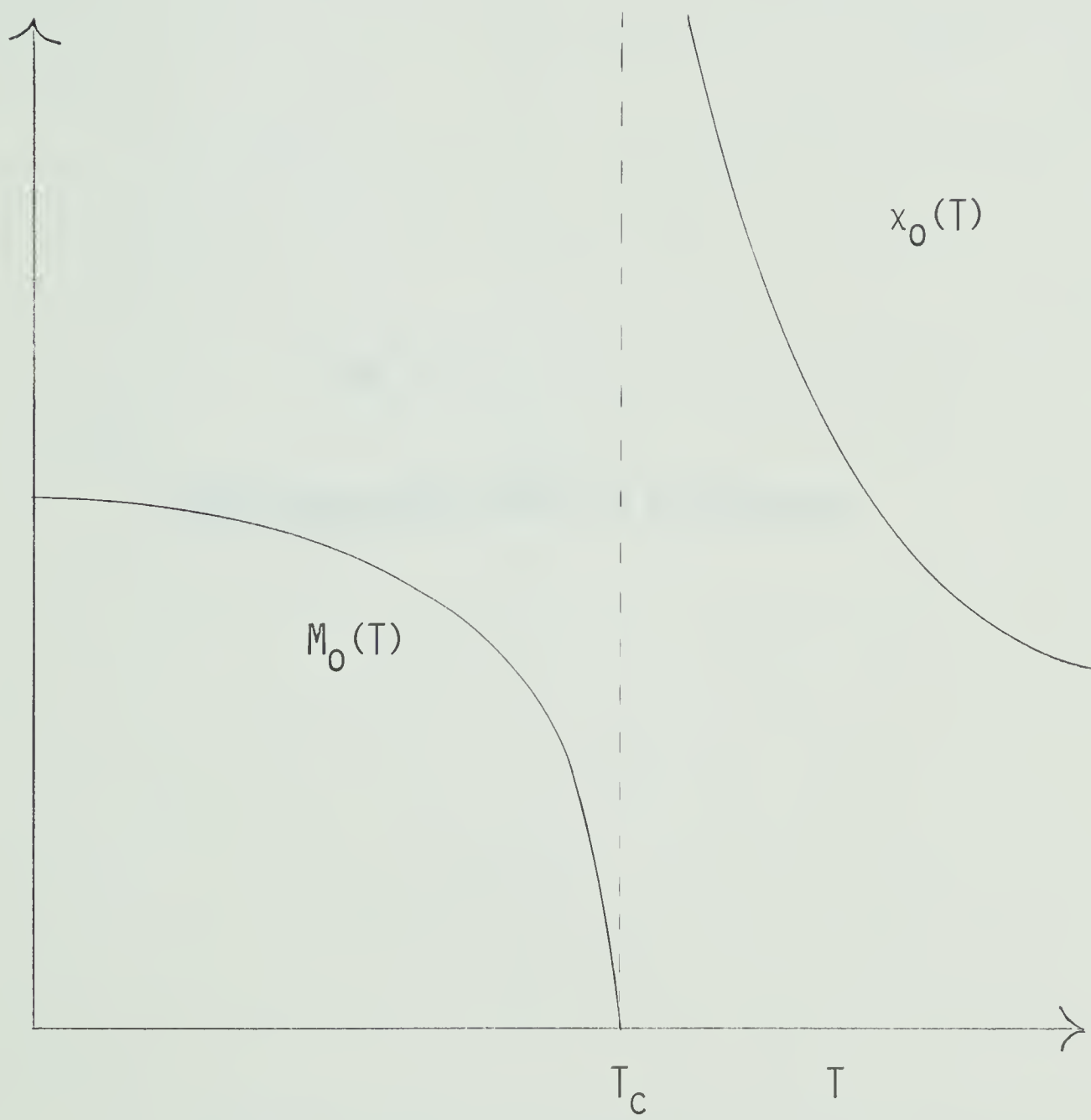
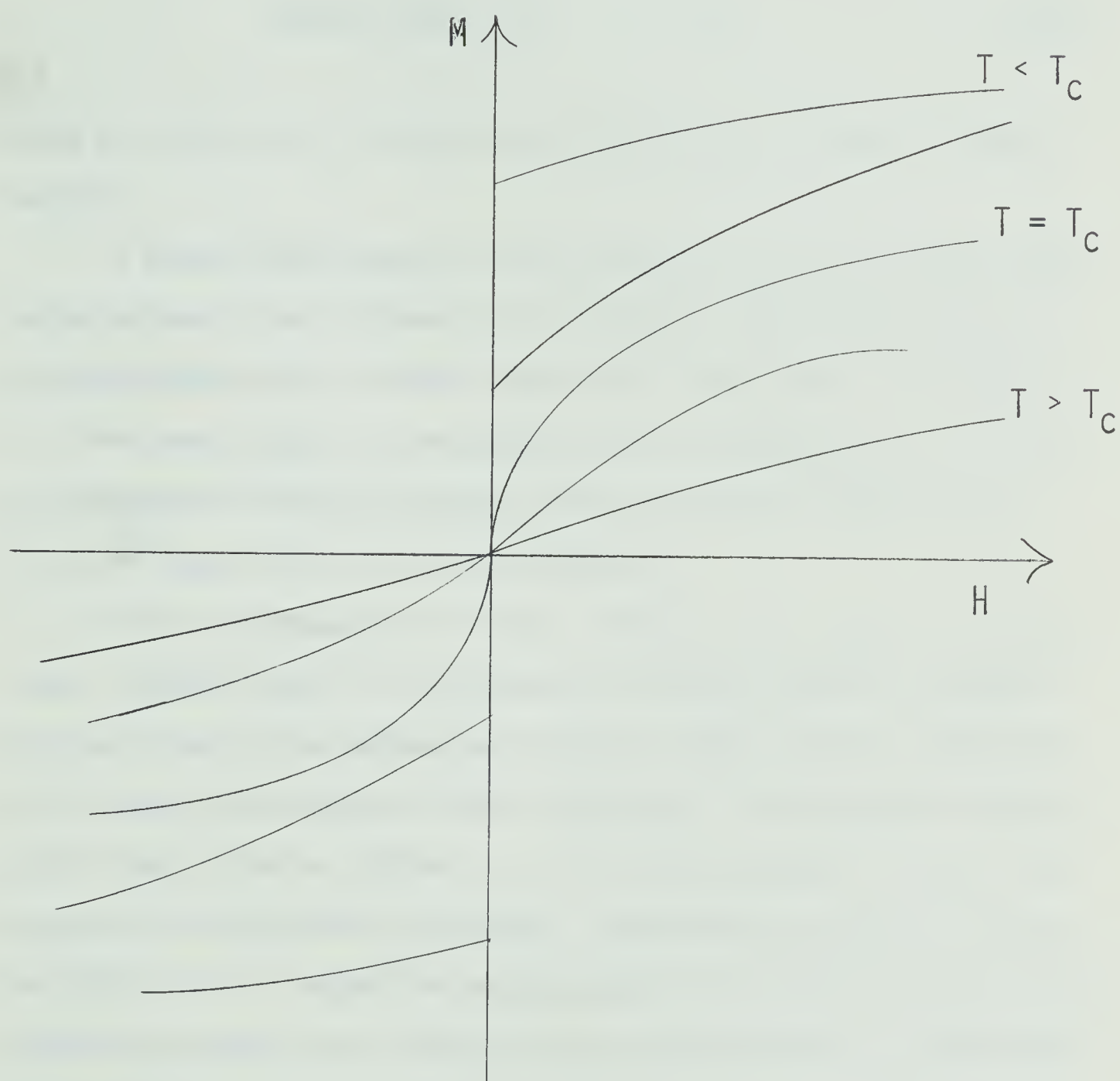


Fig 1.3

Ideal magnetization curve of a ferromagnet



of the insulating ferromagnet EuS and found

$$M_0(T) = A(T_c - T)^\beta \quad (T \rightarrow T_c^-) \quad (1.10)$$

where $\beta = 0.33 \pm 0.01$. A one third law seems also to apply to ferromagnets.

A binary fluid mixture of two components A and B, in which A molecules attract A and B attract B, will undergo a phase separation at low temperatures into an A-rich phase and a B-rich phase. According to J.S. Rowlinson (1959), the coexistence curve, representing in this case the difference in molar fractions of the two phases, again follows a one third power law to a good approximation.

The three systems we have just considered are examples in which 'like' attracts 'like'. The molecules of the gas attract one another, the 'up' spins of the ferromagnet attract up spins and the A molecules of the binary fluid attract other A molecules. If we consider cases in which 'like' attracts "unlike" e.g. the antiferromagnet, we again find significant similarities in behavior. Measurements by Heller and Benedek (1962) of the 'sublattice magnetization' of the antiferromagnet MnF_2 indicate once more that a one third power law describes its temperature dependence.

This discussion could continue with other examples to further emphasize the remarkable similarity in behavior of these seemingly distinct physical systems. However, we shall conclude this section by con-

sidering what all this evidence suggests.

With the possible exception of Bose-Einstein condensation, there is no theory of a phase transition that does not relate the occurrence of the critical point to the existence of some intermolecular interaction. It seems clear the interaction is a necessary prerequisite for a phase transition to take place. One of the most important questions in this field is how important are the details of the interaction in determining the precise behavior of coexistence curves, susceptibilities etc. in the critical region. The experimental evidence we have just reviewed suggests that many of the details are not very important. On the other hand, there must exist some common feature in the interactions present in all these systems that is principally responsible for the observed behavior.

In many respects, the models that theoreticians have examined, particularly the Ising model, are attempts to remove all irrelevant detail from the interactions and calculate the consequences of the one important feature that remains.

We shall return to this question later. But before we become embroiled in the consideration of these details we shall first give a qualitative description of the critical region and why it occurs. Research in this field is dominated by mathematical and computational difficulties as later chapters in this thesis will show and it is important to have a reasonably clear physical picture of critical phenomena before embarking on detailed studies.

B. Description of the Critical Region

Because of the similarities indicated in the last section between the various physical systems exhibiting phase transitions, it is possible to give a qualitative description in a number of different contexts, all of which would be equivalent. One could use the 'language' of simple fluids, of homogeneous binary mixtures or, as we shall choose in this instance, of ferromagnetism and the essential ideas would be the same. Personal preference is dictated largely by one's own background and experience.

We shall consider a simple picture of a ferromagnet in which a large number of 'spins' are located at the sites of a regular lattice. At zero temperature, all the spins are aligned in the same direction (throughout this thesis we are referring to a single domain) giving rise to the spontaneous magnetization. As the temperature rises, excited states of the system are populated in which a gradually increasing number of the spins are reversed in direction (we are assuming in this simplified description that the spins can occupy only two antiparallel orientations). Physical intuition perhaps suggests that the trend from all aligned spins to a random mixture of 'up' spins and 'down' spins should have a smooth temperature dependence in which the final random state is reached in the limit of infinite temperature. Certainly from a theoretical standpoint this is what happens in any finite system. However, this is not what takes place in real ferromagnets or in theoretically infinite

systems (i.e. $N, V \rightarrow \infty$ such that $N/V = \text{const.}$).

A convenient description of what we believe takes place as the temperature increases can be given in terms of the long range order parameter. This is a measure of the tendency of any spin to align itself in the same direction as another spin in the limit that the distance separating the spins becomes infinite. At zero temperature, the long range order is unity. As the temperature rises, it begins to decrease, reflecting the increasing significance of the disrupting effect of the thermal energy of the spins. However, rather than gradually decrease to zero in the infinite temperature limit, the correlation between the two infinitely separated spins becomes abruptly zero at a finite critical temperature. The spontaneous magnetisation per spin is often defined as proportional to the square root of the long range order and so consultation of figure 1.2 indicates how the latter varies from unity at zero temperature to zero at the critical temperature.

We can regard this whole process as a competition between the ferromagnetic interaction and the thermal energy of the spins. As the temperature rises the thermal energy increases and at the critical point it finally and abruptly overcomes the ferromagnetic interaction tending to align the infinitely separated spins.

Let us now approach the critical point from the infinite temperature limit. There, the spins are randomly orientated and behave independently. Thermal energy completely dominates their motion and we say the ferromagnet is behaving as a paramagnet. However, as the temperature

is lowered, the spin-spin interaction begins to make itself felt and short range order gradually becomes significant. By this is meant there exists some degree of correlation between spins which are nearest neighbours on the lattice or not too far separated. If one spin points in a given direction there is a probability greater than one half that its near neighbours will be aligned in the same direction.

As the temperature continues to drop, the correlation length, the maximum separation of appreciably correlated spins, increases until in the critical region it reaches macroscopic dimensions becoming infinite at the critical temperature. The long range order has set in. This increasing correlation between the spins is reflected macroscopically in a rapidly increasing magnetic susceptibility which becomes infinite at the critical point. That the spins should tend to cooperate so rapidly that even at a non-zero temperature two spins infinitely far apart are 'aware' of each other is an indication of how significant is the interaction between the spins. Clearly no non-zero critical temperature could occur in the absence of this interaction. Such phenomena are often referred to as 'cooperative' phenomena and we can see how descriptive the term is.

We hope by calculating the critical behavior of the susceptibility with various types of interaction to determine how much this 'spreading' of the correlation throughout the spin system depends on the details of the interaction. The magnetic susceptibility is therefore an important 'mirror' of the critical behavior and much of this thesis is concerned with its calculation for certain models.

Finally, another interesting feature of the critical region is the existence of large scale fluctuations. Near the critical temperature, there occur correlated deviations of well-separated spins from their equilibrium orientations. These fluctuations are not only spread over large regions of the ferromagnet but persist over long periods of time. They arise because of the delicate balance between the thermal energy and the spin-spin interaction that exists in the critical region. In fluid systems they give rise to critical opalescence and the analogous phenomenon in magnetic systems is the critical scattering of neutrons. There has been much theoretical and experimental activity in this area in recent years (Green and Sengers 1965).

C. Brief Historical Survey

One of the standard treatments of many-body systems and phase transitions which has existed for at least sixty years and is still used at the present time is the 'mean field' or 'internal field' approach. It is an attempt to replace the perhaps complicated pairwise interactions between the particles (or spins) by a uniform internal field with which each particle interacts directly. This field is calculated self-consistently so that the average interaction of each particle with its neighbours is preserved. It represents the first attempt to theoretically predict a phase transition. The Weiss theory of ferromagnetism (1907), the Van der Waals' equation of state (1873) and the Bragg-Williams approximation for binary alloys (1934) are all examples of this approach applied

to different physical systems.

Such theories do succeed in predicting the existence of a critical point but fail in giving the observed behavior of the thermodynamic variables in the critical region. For instance, they predict one half power laws for spontaneous magnetization and coexistence curves which disagree with experimental results. For compressibility and susceptibility, they give the famous Curie-Weiss law, namely,

$$\chi_T = \frac{B_c^+}{|T - T_c|} = \chi_0(T), \quad (T \rightarrow T_{c\pm}) \quad (1.11)$$

which is again not in accord with experiment for temperatures close to critical. The essential reason for their failure is that the very concept of 'mean field' neglects consideration of short-range and long-range order which play such an important role in determining the characteristics of the critical region. It should be mentioned that, in the context of binary alloys, Bethe (1935) attempted to improve on the Bragg-Williams approximation by taking nearest neighbour correlations into account but the improvement in the results was very limited. As one might expect, he did obtain a 'tail' to the specific heat curve above T_c which is not predicted by 'mean-field' arguments. However, the finite discontinuity in the specific heat at T_c , characteristic of the B.-W. approximation, remains.

Practically all modern theories of critical phenomena that have

had any success in comparison with experiment have been based on a very small number of models. The oldest and still most popular is the Ising model introduced and solved exactly in one dimension by Ising (1925). It can be characterized by the Hamiltonian,

$$\mathcal{H} = -J \sum_{(ij)} \sigma_i \sigma_j \quad , \quad (1.12)$$

where i and j label the sites of the lattice, and σ is a two-valued variable (usually chosen to be $+1$ or -1). We shall assume in all our considerations that the summation in (1.12) is over nearest neighbour pairs of sites. This is in direct contrast to the mean field calculations which are equivalent to a system with infinitely long range but infinitely weak interactions. The Ising model will be discussed in more detail in the next section.

Ising's one dimensional solution did not exhibit a non-zero critical temperature, but some years later Peierls (1936) predicted a phase transition could take place in Ising systems of dimension greater than one. Then Kramers and Wannier (1941) deduced the location of the critical temperature for the case of the simple quadratic lattice in zero magnetic field. Finally, Onsager (1944) solved the problem exactly on this lattice and obtained a logarithmic singularity of the specific heat. It is difficult to exaggerate the importance of his solution. Apart from later solutions on other two-dimensional lattices, it

stands alone as an exact solution of a many-body system which exhibits a critical phase transition. It proved conclusively that such critical behavior could be predicted by a model which included only nearest neighbour interactions.

Since then, a great amount of effort has gone into the search for a similar exact solution of the three-dimensional Ising model and has met with singularly little success. Adopting a different approach and developing what has now become known as the 'exact' series expansion technique, the King's College group in London have, however, achieved great success in analysing the behavior of the Ising model and others near their critical temperatures. This approach has been very comprehensively reviewed by Domb (1960). Part I of this thesis is an example of an exact series expansion calculation. These series, be they high temperature or low temperature, are perturbation expansions about infinite or zero temperature in appropriate variables. A finite number of the coefficients (as large as can be calculated within reasonable time limits) are found exactly and from the truncated series, it turns out to be possible to make very accurate predictions of the behavior of these models near T_c . Further details will be given in later chapters.

Another important landmark, particularly with regard to ferromagnetism, was the introduction by Heisenberg (1928) of what is now known as the Heisenberg model of ferromagnetism. It is described by the Hamiltonian,

$$\mathcal{H} = -J \sum_{(ij)} \sigma_i \cdot \sigma_j , \quad (1.13)$$

which differs from (1.12) in that the σ 's are not scalars but quantum mechanical spin operators. It is considered a more realistic model of a ferromagnet than the Ising model but it turns out to be much harder to solve. Indeed, no exact solution in any dimension has been obtained and only approximate techniques such as exact series expansions for high temperatures and spin-wave theory for low temperatures have been successful in predicting the behavior of this model.

D. Discussion of Models

In setting up a theory of a many-body system, some may feel that the ultimate goal is to solve the many-body Schrodinger equation where all contributions to the Hamiltonian have been included. Such a task is not only impossible but quite useless. Once the basic principles of wave-mechanics have been impressively verified by the results of an exact calculation on some very simple system such as the hydrogen atom, it serves little purpose to seek the same degree of accuracy on more and more complicated systems. If that were the only task of theoretical physics, it would indeed be a dull and pedestrian discipline. Rather, when confronted by a complicated many body system, the physicist must use his intuition plus admittedly certain compulsions arising from inevitable mathematical limitations to simplify the problem. He ignores the details he feels are of little relevance to the phenomena he wishes to predict and exaggerates those of primary importance. Comparison of the results of his theory with experiment will ultimately tell him how successful

his 'pruning' has been. In modern physics, our aim is not to produce an exact copy on paper of nature and the above procedure is the only means by which we can improve our understanding.

The commonly used models of critical phenomena, namely the Ising model and the Heisenberg model, can be regarded as rather successful examples of this procedure. The Ising model, in particular, is a case where almost the maximum amount of simplification has taken place. It is difficult to think of any further modifications one could make to (1.12) to simplify it further that does not render the model quite trivial and uninteresting. One frequently reads of how artificial the Ising model is but it is not easy to reconcile this point of view with the great advances in our understanding of phase transitions that have come about from extensive study of its properties. It can be used as a model of any system involving lattice sites with two states per site. The lattice gas model in which the volume of the gas is divided into cells which can either be empty or occupied by one gas molecule, the ferromagnetic and antiferromagnetic in which each site has either an 'up' spin or a 'down' spin and the binary alloy with either A or B type molecules at the sites are all examples to which the Ising model can readily be applied. In terms of this model, then, we can understand the similarity between the different physical systems outlined in section A.

As an example of the predictions of the model, the index ' β ' describing the behavior of the coexistence and magnetization curves in the region of the critical point is predicted by three dimensional Ising

models to be given by (Fisher and Essam 1963),

$$\beta = 0.312 \pm 0.002, \quad (1.14)$$

which is remarkably close to the experimental value of almost one third quoted in section A. This result strongly indicates that the model incorporates most of the essential features determining the nature of the critical region, but the small discrepancy with experiment is significant. One of the outstanding theoretical questions still remaining is exactly what has been omitted in setting up the Ising model Hamiltonian.

One could go on and describe many of the other interesting results given by the model. However, we shall summarize by noting that in the past the vast amount of work done was concerned with equilibrium properties such as specific heat, susceptibility or compressibility and nature of coexistence curves. Recently there has been a shift in emphasis to a study of the non-equilibrium or dynamic properties of the model. Discussions of critical point scattering by Fisher and Burford (1967) and isothermal frequency dependent susceptibility by Glauber (1963) are examples.

The second part of this thesis is an example of such a study. We determine exactly for two-dimensional Ising models the frequency dependent perpendicular susceptibility (in the context of magnetism a measure of the response of the system to a magnetic field applied perpendicular to the direction along which the spins can align) and also the in-

elastic neutron scattering cross-section. Just as the equilibrium properties of the model have so closely mirrored the properties of real systems, we hope that such studies of the dynamical features of the model will lead to further understanding of critical phenomena.

Thus far, in our discussion of models, we have concentrated on the Ising model since it is the 'prototype' of all such models. However, particularly with regard to ferromagnetism, the Heisenberg model is at least equally important. Its physical origin lies in the quantum mechanical exchange forces that exist between systems of electrons, and its discovery by Heisenberg (1928) provided physicists with a much sought-after source of ferromagnetism. A simple derivation of the model following closely Heisenberg's original arguments will be given in the next chapter. This will lead naturally to the introduction of the spin one exchange model of ferromagnetism the study of which forms the first part to this thesis. The introduction of this model is an attempt to extend the very limited number of models which have been investigated. One of the principal aims is to shed some more light on the question that has been mentioned several times throughout this introduction i.e. how important is the nature of the interaction in determining the characteristics of the critical region.

Chapter three contains a number of preliminaries to the main calculation, namely an introduction to high temperature series expansions and some discussion of graph theory. Detailed consideration is given in chapter four to the two main techniques of evaluating exact series expan-

sions, the direct method and the linked cluster method, and the application of the latter to the spin one exchange model is described. Chapter five discusses the group theoretic techniques employed to simplify the calculation and part one of the thesis concludes with detailed analysis and discussion of the results in chapter six.

Part two begins with an introduction to the concept of the perpendicular susceptibility of the Ising model and how it can yield dynamic features of the model. Also in chapter seven, the Kubo formalism on which the whole calculation is based is discussed in general terms and then applied to the problem of the perpendicular susceptibility. The next chapter contains some discussion of the interesting exact results obtained for the plane square and honeycomb lattices. The resolution of some apparent discrepancies is then described in chapter nine. In chapter ten, the same techniques used in this calculation are applied to determine the inelastic neutron scattering cross-section of the Ising model. Finally in chapter eleven a general survey of the results obtained in both parts of the thesis is given.

PART I

THE SPIN ONE EXCHANGE
MODEL OF FERROMAGNETISM

CHAPTER II

THE EXCHANGE INTERACTION

We shall consider a quantum mechanical system of two identical particles (labelled 1 and 2) of spin S with an interaction potential V_{12} which is symmetric under exchange of the two particles. Our aim is to develop an expression for what is known as the exchange interaction particularly for the cases $S = \frac{1}{2}$ and 1, though all our arguments can easily be extended to higher spin.

We introduce the operator P_{12} which represents the exchange of the two particles. It has two eigenvalues, +1 for symmetric states and -1 for antisymmetric states. Since both total spin $\underline{S}_{12} = \underline{S}_1 + \underline{S}_2$ and total z component of spin $S_{12}^z = S_1^z + S_2^z$ commute with P_{12} , we can conclude that any simultaneous eigenstate of \underline{S}_{12} and S_{12}^z has a definite symmetry (i.e. is an eigenstate of P_{12}). Indeed any non-degenerate eigenfunction of a commuting set of symmetric operators will likewise be either symmetric or antisymmetric. In addition, since P_{12} commutes with $S_{12}^{\pm} = (S_1^x + S_2^x) \pm i(S_1^y + S_2^y)$, the symmetry character of the simultaneous eigenstate is independent of S_{12}^z and is determined solely by the value of \underline{S}_{12} . This means that any eigenstate of \underline{S}_{12} even if it is not also an eigenstate of S_{12}^z will have a definite symmetry since it can be considered as a linear combination of the simultaneous eigenstates of \underline{S}_{12} and S_{12}^z . Such a linear combination will have the same

symmetry character as each of its components.

One can also show that, as the total spin quantum number S_{12} decreases from 2 to 0, the corresponding sets of eigenstates alternate in symmetry character with the '2S' states always being symmetric. A simple proof due to J. Hamilton is contained in a paper on the general connection between exchange and spin by Schrodinger (1941). We shall illustrate it for the case of spin 1 particles.

We try by inspection to write down systematically all the eigenstates of S_{12}^Z of definite symmetry. Then by a process of elimination, we determine which eigenvalue of S_{12} belongs to that state and hence the symmetry character associated with that eigenvalue. Let $\alpha(i)$, $\beta(i)$, $\gamma(i)$ represent the eigenstates of S_i^Z belonging to the eigenvalues 1, 0, -1 respectively. For the case of $S_{12}^Z = 2$, there is only one possible eigenstate of definite symmetry, namely $\alpha(1)\alpha(2)$. This state must also have $S_{12} = 2$ and therefore all eigenstates of S_{12} with $S_{12} = 2$ are symmetric. For $S_{12}^Z = 1$, we can have either $\alpha(1)\beta(2) + \alpha(2)\beta(1)$ or $\alpha(1)\beta(2) - \alpha(2)\beta(1)$. The former must be associated with $S_{12} = 2$ since as we have said, all such states are symmetric and hence the latter is a state whose total spin quantum number is 1. We conclude that all states with $S_{12} = 1$ are antisymmetric. When $S_{12}^Z = 0$, there exist three possibilities viz. $\alpha(1)\gamma(2) + \alpha(2)\gamma(1)$, $\alpha(1)\gamma(2) - \alpha(2)\gamma(1)$ and $\beta(1)\beta(2)$ i.e. two symmetric and one antisymmetric states. Now one symmetric state must be associated with $S_{12} = 2$, the antisymmetric state with $S_{12} = 1$ and hence the state with $S_{12} = 0$ must be symmetric. Thus as we

stated earlier, the symmetry character alternates from $S_{12} = 2$ to $S_{12} = 0$. For the spin $\frac{1}{2}$ case, we would similarly find that $S_{12} = 1$ states are symmetric (the triplet states) and for $S_{12} = 0$, the state is antisymmetric (the singlet state). This conclusion that the symmetry character and total spin quantum number of an eigenstate are closely linked forms an important step in the argument that Heisenberg (1928) used to derive an expression for the exchange operator P_{12} . Our derivation will closely follow an account given by Van Vleck (1932).

Let us first suppose that the two particles do not interact and for the moment we do not consider spin. The Schrodinger wave equation for the system then is

$$\nabla_1^2 \psi(1) + \nabla_2^2 \psi(2) + \frac{8\pi^2 m}{h^2} [W - V(1) - V(2)] \psi = 0 \quad (2.1)$$

where m is the mass of each particle and $V(i)$ is the potential to which the i th particle is subjected independently of the presence of the other particle. The solutions of this equation are of course of the form,

$$\psi_I(1,2) = \psi_k(1)\psi_m(2), \quad (2.2)$$

associated with the energy eigenvalue $W = W_k + W_m$ where ψ_k and ψ_m are solutions of the single particle Schrodinger equation belonging to the eigenvalues W_k and W_m respectively. An alternative solution, however, exists associated with the same eigenvalue, namely,

$$\psi_{II}(12) = \psi_k(2)\psi_m(1) , \quad (2.3)$$

where (2.3) has been obtained from (2.2) by exchanging the two particles. This exchange degeneracy is removed when a symmetric interaction between two particles V_{12} is introduced. It is a simple exercise to show that the two eigenstates now become

$$\psi_{\text{sym}} = \frac{1}{\sqrt{2}} (\psi_I + \psi_{II}) \quad (2.4)$$

and

$$\psi_{\text{ant}} = \frac{1}{\sqrt{2}} (\psi_I - \psi_{II}) \quad (2.5)$$

associated respectively with the eigenvalues,

$$W = \begin{matrix} W_0 + K_{12} + J_{12} & \text{(a)} \\ W_0 + K_{12} - J_{12} & \text{(b)} \end{matrix} \quad (2.6)$$

where W_0 is the energy in absence of the interaction term V_{12} ,

$$K_{12} = \iint \psi_I V_{12} \psi_I dv_1 dv_2 = \iint \psi_{II} V_{12} \psi_{II} dv_1 dv_2 \quad (2.7)$$

and

$$J_{12} = \iint \psi_I V_{12} \psi_{II} dv_1 dv_2 \quad (2.8)$$

We now consider the inclusion of spin. For spin $\frac{1}{2}$ particles, the Pauli exclusion principle demands that the total wave function be antisymmetric under exchange of the two particles. In the absence of any coupling between space variables and spin variables, the total wave function will be the product of a space dependent part and a spin dependent part. Then the spin wave function associated with (2.4) must be antisymmetric and the one associated with (2.5) must be symmetric i.e. the eigenvalues of P_{12} are -1 and +1 respectively where now P_{12} represents the exchange of only the spins of the particles. In other words, P_{12} has eigenvalues +1 and -1 for states where, from (2.6), V_{12} has eigenvalues $K_{12} - J_{12}$ and $K_{12} + J_{12}$. We can therefore set up the important operator identity,

$$V_{12} = K_{12} - J_{12}P_{12} \quad (2.9)$$

Heisenberg's principal achievement was essentially to demonstrate (2.9) for the case of electrons where V_{12} is the familiar coulomb interaction. We should emphasize that this exchange operator is simply a manifestation in spin space of the inter-electron coulomb repulsion which comes about principally because of the Pauli exclusion principle. The often used phrase that the exchange interaction has no classical analogue is therefore a little misleading.

The spin exchange operator P_{12} can be put in a much more familiar form. For the antisymmetric spin state of two spin $\frac{1}{2}$ particles, we

found that S_{12} must equal 0. As a consequence the operator $\underline{S}_1 \cdot \underline{S}_2$ given by,

$$\underline{S}_1 \cdot \underline{S}_2 = \frac{1}{2} (\underline{S}_{12}^2 - \underline{S}_1^2 - \underline{S}_2^2) \quad (2.10)$$

$$= \frac{1}{2} (\underline{S}_{12}^2 - \frac{3}{2}) , \quad (2.11)$$

must have an eigenvalue of $-\frac{3}{4}$ in that state. Similarly for the symmetric state, $\underline{S}_1 \cdot \underline{S}_2$ has an eigenvalue of $\frac{1}{4}$. It is therefore possible to set up another important operator identity of the form,

$$P_{12} = a + b \underline{S}_1 \cdot \underline{S}_2 , \quad (2.12)$$

where a and b are constants determined by the simultaneous equations ,

$$\left. \begin{aligned} -1 &= a - \frac{3}{4} b \\ 1 &= a + \frac{1}{4} b \end{aligned} \right\} . \quad (2.13)$$

The solution yields the familiar Dirac relation ,

$$P_{12} = \frac{1}{2} + 2 \underline{S}_1 \cdot \underline{S}_2 . \quad (2.14)$$

It is in this form that the Heisenberg exchange interaction is usually written i.e. neglecting a relatively unimportant constant,

$$V_{12} = -JS_1 \cdot S_2 \quad . \quad (2.15)$$

The purpose of this detailed discussion has been not only to emphasize the physical origins of exchange but to introduce the spin one exchange operator which is obtained by a natural extension of equations (2.9) to (2.15) to the spin one case. For the symmetric spin state, S_{12} must be 2 or 0 and 1 for the antisymmetric state. Proposing for P_{12} a form,

$$P_{12} = a(\underline{S}_1 \cdot \underline{S}_2)^2 + b(\underline{S}_1 \cdot \underline{S}_2) + c \quad , \quad (2.16)$$

we obtain the operator identity ,

$$P_{12} = (\underline{S}_1 \cdot \underline{S}_2)^2 + (\underline{S}_1 \cdot \underline{S}_2) - 1 \quad , \quad (2.17)$$

and hence a spin one exchange interaction which can be written in the form ,

$$V_{12} = -J[(\underline{S}_1 \cdot \underline{S}_2)^2 + (\underline{S}_1 \cdot \underline{S}_2) - 1] \quad . \quad (2.18)$$

A model where the basic interaction is given by (2.18) will be investigated in the following chapters.

The Heisenberg exchange given by (2.15) has also been investigated for higher spin systems. In these cases, it represents the sum of all the single electron exchanges between the electrons in the spin system with the assumption that the interaction constant, J , is the

same for all such exchanges. For a spin 1 system (2.18) represents the simultaneous exchange of the system of electrons in a relative spin 1 state. In other words, if the electrons are so coupled that they can be considered as a single spin 1 system rather than as a system of independent electrons, we might expect (2.18) to be significant.

There are other important reasons for considering a spin 1 exchange model. For spin 1 systems, the most general form of isotropic spin-spin interaction is given by (2.16). Since $\underline{S}_1 \cdot \underline{S}_2$ has only three distinct eigenvalues, it is easy to show that $(\underline{S}_1 \cdot \underline{S}_2)^n$ for $n > 2$ can be expressed in the form (2.16). While such interactions have not been considered previously by theoreticians in this field, it should be interesting to see what the most general form of interaction for a spin 1 system predicts particularly in comparison to the spin 1 Heisenberg model. The reason that the particular form (2.17) is chosen is partly for arguments given in the previous paragraph and partly since as we shall find later, the spin 1 exchange operator allows for considerable mathematical simplification. As we have said earlier, we can also hope to shed some light on the question of how important is the nature of the interaction in determining the details of a phase transition.

Finally, let us mention an important case where an interaction of the form (2.16) arises. In spin notation, the components $Q^{ij}(i,j=x,y,z)$ of the quadrupole moment tensor are given by,

$$Q^{ij} = 3 \frac{S^i S^j + S^j S^i}{2} - \delta_{ij} S^2 \quad . \quad (2.19)$$

Segré (1953) has shown that a quadrupole-quadrupole interaction of the form ,

$$V_{12} = J \sum_{ij} Q_1^{ij} Q_2^{ij} , \quad (2.20)$$

can be written as,

$$V_{12} = J \left[9(\underline{s}_1 \cdot \underline{s}_2)^2 + \frac{9}{2} (\underline{s}_1 \cdot \underline{s}_2) - 3\underline{s}_1^2 \underline{s}_2^2 \right] . \quad (2.21)$$

Now Nakamura (1955) has argued that the principal interaction in solid hydrogen with a high concentration of ortho-molecules (> 60%) is an electric quadrupole-quadrupole interaction where the interaction constant, J , is given explicitly by,

$$J = \frac{6e^2 Q_{H_2}^2}{25 R^5} . \quad (2.22)$$

'e' is the electronic charge, Q_{H_2} the quadrupole moment of the ortho-hydrogen molecule and R is the distance between interacting quadrupoles. Using data given by Nakamura, we find

$$J = 0.654k^0K \quad (2.23)$$

where k is Boltzmann's constant. That the interaction constant can be evaluated is in direct contrast with most magnetic systems. Another feature worth noting is that the $\frac{1}{R^5}$ dependence of J would make the

normal nearest neighbour interaction approximation a good one in this instance.

While, of course, (2.21) does not have its physical origins in exchange, it is nevertheless formally an example of (2.16). Unfortunately for statistical-mechanical calculations, the interaction constant, as we have seen from (2.22), is positive and leads to an 'antiferromagnetic' type of interaction. The case we shall investigate in the following chapters is the one where the ferromagnetic state is preferred. Experience has shown that series expansion techniques give more accurate results for ferromagnetic systems.

CHAPTER III

AN INTRODUCTION TO HIGH TEMPERATURE SERIES EXPANSIONSA. Review of Statistical Mechanics

As we have just indicated in the previous chapter, the spin one exchange model of ferromagnetism is characterized by the Hamiltonian,

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1 \quad (3.1)$$

$$= -J \sum_{n,n} \left[(\underline{S}_i \cdot \underline{S}_j)^2 + (\underline{S}_i \cdot \underline{S}_j) - \frac{4}{3} \right] - mH \sum_i S_i^z, \quad (3.2)$$

where m is the magnetic moment associated with each spin and H represents an external magnetic field aligned along the z direction of our coordinate system. In \mathcal{H}_0 , i and j label the sites of a regular lattice (in our case we shall consider the face-centred cubic lattice) and the summation is taken over all nearest neighbour pairs of sites. The interaction constant J is considered positive so that the system is ferromagnetic. The interaction differs by a constant from that introduced in (2.18). This alteration has no effect on the final results but it does introduce significant simplification into the calculation by making the interaction matrix traceless. \mathcal{H}_1 represents the interaction of the system with an external magnetic field. We shall always consider the

limiting case of this field tending to zero.

Our aim is to proceed from the microscopic description of our system as given by (3.2) to calculate the familiar macroscopic thermodynamic variables such as specific heat, entropy and magnetic susceptibility. The whole formalism of statistical mechanics is set up primarily for this purpose. For systems with a fixed number of particles N , in thermal contact with a heat bath, the function which provides this important bridge between the microscopic and the macroscopic is the canonical partition function given for quantum mechanical systems by,

$$Z(T,H) = \text{trace} [\exp(-\beta \mathcal{H})] , \quad (3.3)$$

where $\beta = 1/kT$ (k is Boltzmann's constant) and \mathcal{H} is the Hamiltonian matrix operator given for our model by (3.2). The fundamental equation in statistical mechanics links the partition function to the Helmholtz free energy F , namely,

$$F = -kT \log Z . \quad (3.4)$$

Simple thermodynamics then gives for the entropy S ,

$$S = \frac{\partial}{\partial T} (kT \log Z) , \quad (3.5)$$

for the magnetic specific heat C ,

$$C = \frac{\partial}{\partial T} \left[kT^2 \frac{\partial}{\partial T} (\log Z) \right], \quad (3.6)$$

and for the magnetic susceptibility χ ,

$$\chi = \frac{\partial^2}{\partial H^2} (kT \log Z) . \quad (3.7)$$

It should be noted that in (3.5-7) no indication is given of whether the derivatives are taken at constant magnetic field or constant magnetization. We shall always be considering the $H = 0$ limit which for a ferromagnetic in the high temperature region above the critical point is identical with the $M=0$ condition.

As mentioned in chapter I, in the high temperature series expansion method, (3.3) is expanded as a power series in inverse temperature. If we introduce the convenient notations,

$$\left. \begin{aligned} P &= \sum_{nn} \left[(\underline{S}_i \cdot \underline{S}_j)^2 + (\underline{S}_i \cdot \underline{S}_j) - \frac{4}{3} \right], \\ Q &= \sum_i S_i^z, \\ \alpha &= \frac{mH}{J}, \end{aligned} \right\} \quad (3.8)$$

substitute (3.2) in (3.3) and expand the exponential, we obtain

$$Z(T,H) = \sum_{r=0}^{\infty} \frac{\text{trace}(P + \alpha Q)^r}{r!} K^r \quad \left[K = \frac{J}{kT} \right] \quad (3.9)$$

$$= \text{trace } I + \sum_{r=1}^{\infty} \frac{\text{trace}(P + \alpha Q)^r}{r!} K^r . \quad (3.10)$$

I is the unit matrix which for a system of N spin 1 particles has dimensions $3^N \times 3^N$. (3.10) can then be written in the form,

$$Z(T,H) = 3^N \left(1 + \sum_{r=1}^{\infty} \frac{\mu_r}{r!} K^r \right) \quad (3.11)$$

where

$$\mu_r = \frac{\text{trace}(P + \alpha Q)^r}{3^N} . \quad (3.12)$$

To obtain the entropy etc., we are primarily interested in $\log Z(T,H)$ which from (3.11) is given by,

$$\log Z(T,H) = N \log 3 + \log \left[1 + \sum_{r=1}^{\infty} \frac{\mu_r}{r!} K^r \right] \quad (3.13)$$

$$= N \log 3 + \sum_{r=1}^{\infty} \frac{\lambda_r}{r!} K^r . \quad (3.14)$$

In the language of statistics, the coefficients λ_r are the cumulants

corresponding to the moments μ_r . Explicit relations giving λ_r as combinations of all the μ_s with $s \leq r$ are to be found in appendix A for $r \leq 9$ and for the particular case of $\mu_1 = 0$. Alternatively the λ_r can be regarded as those parts of the μ_r that are directly proportional to N . Since $\log Z$ is an extensive quantity, λ_r must be proportional to N and, in fact,

$$\mu_r = \lambda_r + \text{terms proportional to } N^2, N^3 \text{ etc.}, \quad (3.15)$$

these latter terms cancelling in the transformation from (3.13) to (3.14).

For the initial magnetic susceptibility, χ_0 , using (3.7), (3.8) and (3.14), we obtain

$$\chi_0 = \frac{kTm^2}{J^2} \sum_{r=1}^{\infty} \frac{\lambda_r''}{r!} K^r \quad (3.15)$$

where

$$\lambda_r''(\alpha) = \lim_{\alpha \rightarrow 0} \frac{\partial^2 \lambda_r(\alpha)}{\partial \alpha^2} \quad (3.16)$$

From (3.8), (3.12) and appendix A, we find

$$\lambda_1 = \mu_1 = 0 = \lambda_1'' \quad (3.17)$$

and, as a result, (3.15) is frequently written in terms of the reduced

initial susceptibility, $\overline{\chi}_0$, given by,

$$\overline{\chi}_0 = \frac{kT}{m^2} \chi_0 = \sum_{r=2}^{\infty} \frac{\lambda_r''}{r!} K^{r-2} . \quad (3.18)$$

Again λ_r'' can either be regarded as that part of μ_r'' [μ_r'' is defined in an analogous manner to λ_r''] which is proportional to N or as a particular combination of all the μ_s and μ_s'' for $s \leq r$. These combinations are also given in appendix A for $r \leq q$.

The object of this technique is to calculate the exact numerical value of as many of the λ_r and λ_r'' as possible. The former will give the series for the entropy, specific heat and internal energy in zero external field (if we put $\alpha=0$ in (3.12)) and the latter the initial magnetic susceptibility. The first term in each case reproduces the behavior in the infinite temperature limit. As more terms are evaluated, the series will represent a good approximation to the exact behavior at lower and lower temperatures. Ideally to obtain information close to the critical temperature, we should simply calculate a sufficient number of terms of the series. However, the great labour involved in finding these coefficients strictly limits the number which it is possible to calculate. Fortunately there exist techniques which enable us to predict the behavior of the series near the critical temperature when the truncated series by itself would represent a bad approximation.

First, however, we shall describe the two important and distinct

methods that are used to calculate the coefficients. They are known as the direct method and the cluster expansion method. In both cases, an integral part of the procedure is to set up a correspondence between the many traces of products of spin variables that contribute to any given λ_r , μ_r etc. and various types of linear graphs. As a necessary preliminary, we shall give precise definitions of many of the terms that now form part of the established 'vernacular' of graph theory. We shall follow very closely those given by Sykes et al. (1966) who in turn used Ore (1962) as their principal reference.

B. Concepts in Graph Theory

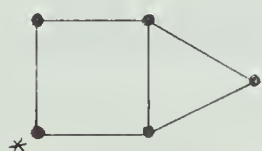
A linear graph consists of a set of vertices (points) V , and associated pairings of these vertices. These pairings are represented by the lines (edges or bonds) of the graph. An undirected graph is one in which the order of the vertices in a pair is not considered. We shall be concerned throughout solely with undirected graphs.

A graph is connected if for every pair of vertices there is at least one sequence of lines called a path such that the first line is incident upon one of the vertices and the last line incident on the other, consecutive lines always having a common vertex and no line being repeated in the sequence. A disconnected graph is one in which there exists at least one pair of vertices between which there is no such path. The graph drawn below is an example of a disconnected undirected linear graph of three connected components for which six vertices constitute

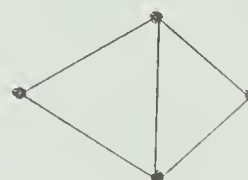
the vertex set and four edges the edge set.



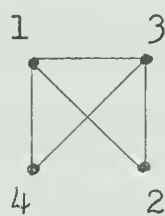
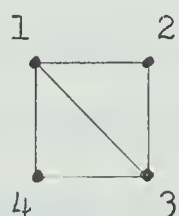
The degree (or order) of a vertex is equal to the number of lines which are incident on that vertex. The above graph has three vertices of order two, two of order one and one of zero order. Vertices of order greater than two are known as nodes while those of degree two or less are called antinodes. A bridge is a simple path connecting two nodes, a simple path being one which traverses a given vertex no more than once. The suppression of a vertex of degree two is defined as the removal of the vertex and the identification of the two lines incident upon it. Conversely the insertion of a vertex of degree two is the operation of replacing a line incident upon two such vertices by two lines and one vertex, each line incident upon one of the original vertices and the two lines together incident upon the new vertex. The two operations are illustrated below with the vertex in question marked by an asterisk.



↗ INSERTION
 —————
 SUPPRESSION ↘



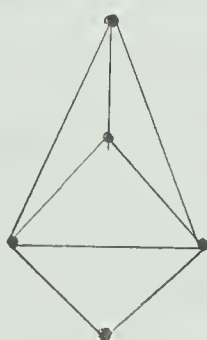
Two graphs are isomorphic if there is a one to one correspondence between their vertices such that two vertices are joined by a line in one of them if and only if they are joined by a line in the other. The two graphs drawn below are isomorphic and the vertices are labelled to show the one to one correspondence.



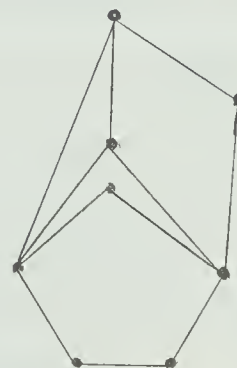
Two graphs are said to be homeomorphic if one of them may be derived from the other by the insertion or suppression of any number of vertices of degree two. Such graphs are said to be of the same topological type or have the same topology. Each graph is a realisation of the topology. For example, (a) is the schematic representation of a certain topology and (b) and (c) are two of the infinite number of distinct realisations of that topology.



(a)



(b)



(c)

If we denote by n , the number of connected components of a graph, e the number of lines and v , the number of vertices, then we define the cyclomatic index c , of the graph to be

$$c = e - v + n . \quad (3.19)$$

This is a very important concept in the classification of graphs. It should be noted that all homeomorphic graphs have the same cyclomatic index. For example all connected linear chains have cyclomatic index zero.

If g_r and g_s are two graphs with disjoint vertex sets and disjoint edge sets, we denote by $g_r \cup g_s$ the graph whose vertex and edge sets are the unions of the respective sets of g_r and g_s . For example,



The graph H is a subgraph of a graph G when the vertex set $V(H)$ is contained in the vertex set $V(G)$ and all the edges of H are edges of G . A graph H is a section graph of G if the vertex set $V(H)$ is contained in the vertex set $V(G)$ and if the lines of H are all the lines of G which connect the vertices of H . For example (b) below is both a subgraph and a section graph of (a) but (c) is only a subgraph.



(a)



(b)



(c)

Any subgraph of G that is isomorphic with a graph g is said to represent a weak embedding of g in G . Likewise any section graph of G which is isomorphic with g is said to represent a strong embedding of g in G . Clearly a strong embedding is also a weak embedding but not vice versa. The lattice constant of a graph g is defined in two senses. The weak lattice constant of g on a graph G , denoted by $(g;G)$, is equal to the number of subgraphs of G which are isomorphic with g . Correspondingly, the strong lattice constant of g on G , denoted by $[g;G]$ is the number of section graphs of G isomorphic with g . For example

$$\left(\begin{array}{c} \bullet \\ \diagup \quad \diagdown \\ \bullet \quad \bullet \end{array} ; \begin{array}{c} \bullet \\ \diagup \quad \diagdown \\ \bullet \quad \bullet \\ \hline \bullet \quad \bullet \end{array} \right) = 3$$

$$\left[\begin{array}{c} \bullet \\ \diagup \quad \diagdown \\ \bullet \quad \bullet \end{array} ; \begin{array}{c} \bullet \\ \diagup \quad \diagdown \\ \bullet \quad \bullet \\ \hline \bullet \quad \bullet \end{array} \right] = 0$$

$$\left(\begin{array}{c} \bullet \\ \diagup \quad \diagdown \\ \bullet \quad \bullet \end{array} ; \begin{array}{c} \bullet \quad \bullet \\ \diagup \quad \diagdown \\ \bullet \quad \bullet \\ \hline \bullet \quad \bullet \end{array} \right) = 8$$

$$\left[\begin{array}{c} \bullet \\ \diagup \quad \diagdown \\ \bullet \quad \bullet \end{array} ; \begin{array}{c} \bullet \quad \bullet \\ \diagup \quad \diagdown \\ \bullet \quad \bullet \\ \hline \bullet \quad \bullet \end{array} \right] = 2$$

It turns out that the strong lattice constants arise in low tem-

perature calculations. In our case, we shall be concerned only with the weak lattice constants and in succeeding chapters this will always be implied even if specific reference to 'weak' is not made.

It is necessary to generalise the concept of lattice constant to the case when G is an infinite lattice. Clearly as defined the lattice constant for this case would be either zero or infinity. If we consider first a lattice of N sites and ignore boundary effects, the lattice constant can be expressed as a function of N . Connected graphs have lattice constants proportional to N and since for the most part, we shall be concerned with connected graphs we redefine lattice constant to mean the number of embeddings per site when the term is applied to an infinite lattice.

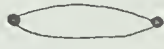

To conclude these introductory remarks on some of the relevant concepts of graph theory, we emphasize that thus far it has been implicitly assumed that a pair of vertices could be connected by no more than one edge. The graph concept can be extended to allow a pair of vertices to be joined by several distinct edges. This is particularly important to our discussion of the 'direct' method of series analysis. This together with our calculation using the 'cluster expansion' method will be discussed in detail in the next chapter.

CHAPTER IV

THE EVALUATION OF SERIES COEFFICIENTSA. The Direct Method

It is clear from (3.8) and (3.10) that in order to calculate the high temperature series for the zero field partition function, we have to evaluate

$$\text{trace}(P^n) = \text{trace} \left[\sum_{n,n.} (P_{ij} - \frac{1}{3}) \right]^n. \quad (4.1)$$

It is very convenient to set up a correspondence between the products of spin operators that contribute to (4.1) for a given 'n' and sets of linear graphs of n bonds. The graphs are unlabelled and the particular sites to which the spin operators are attached is of no relevance. Only the topology of the spin products is important. Thus for n=2, both $(P_{12} - 1/3)^2$ and $(P_{23} - 1/3)^2$ are represented by the graph  and $(P_{12} - 1/3)(P_{23} - 1/3)$ will be represented by the graph  whatever the angle between the bonds (12) and (23) on the lattice under consideration. There are then three main steps in the calculation, namely

a) for a given 'n', to enumerate all possible linear graphs consisting of n bonds,

b) to calculate the lattice constants of these graphs and

c) to evaluate the traces of all the products of spin variables corresponding to each graph.

The enumeration of graphs has been considerably simplified by our modification of the nearest neighbour interaction from P_{ij} to $P_{ij} - 1/3$. In the latter form, the following rearrangement is possible, namely,

$$P_{ij} - \frac{1}{3} = \sum_{m=1}^{12} a_i^m a_j^m, \quad (4.2)$$

where the a^m ($m = 1, 2, \dots, 12$) are all traceless matrices. (They are listed in appendix B.) The Heisenberg interaction is of this form where 'a' is the spin operator S and $m = x, y$ and z .

The spin space of a collection of spins is the direct product of the individual spin spaces. Any product of spin operators belonging to different lattice sites is actually a direct product of the corresponding spin matrices. For example, any term from (4.2) when written in a matrix representation takes the form,

$$a_1^m a_2^m = (a_1^m \times I) \cdot (I \times a_2^m) \quad (4.3)$$

$$= (a_1^m \cdot I) \times (I \cdot a_2^m) \quad (4.4)$$

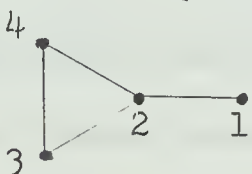
$$= a_1^m \times a_2^m, \quad (4.5)$$

where on the right hand side of (4.3-5) the '.' refers to ordinary matrix multiplication and the 'x' to direct product multiplication.

From a well known property of the trace we can immediately conclude that

$$\text{trace}(a_1^m \times a_2^m) = (\text{trace } a_1^m)(\text{trace } a_2^m). \quad (4.6)$$

(4.3-6) can easily be extended to the general case of a product of any number of spin variables operating on different lattice sites. Since the a's have all be constructed to be traceless, these results have the important consequence that any product involving only one 'a^m' on at least one lattice site will automatically be traceless. Such a situation arises in all spin products corresponding to graphs with at least one vertex of order one. As an example, in P⁴ a typical product cor-

responding to the graph  is given by,

$$\text{trace}(a_1^6 a_2^6 a_2^3 a_2^5 a_4^3 a_4^9 a_3^9 a_3^5) \quad (4.7)$$

$$= (\text{trace } a_1^6)(\text{trace } a_2^6 a_2^3 a_2^5)(\text{trace } a_4^3 a_4^9)(\text{trace } a_3^9 a_3^5)$$

$$= 0, \quad (4.8)$$

$$\text{since } \text{trace } a_1^6 = 0. \quad (4.9)$$

We can therefore ignore all such graphs (often referred to as open graphs or configurations) in our enumeration and this represents an important simplification. Nevertheless the number of graphs becomes quite large as the number of bonds increases. Rushbrooke and Wood (1958) using this method on the Heisenberg model report that, even with further simplifications they were able to introduce, they enumerated a total of 120 graphs with a maximum of six bonds of which 91 were disconnected.

In enumerating these graphs no account is taken of the specific lattice structure. The features of a given lattice arise in the calculation of the lattice constants. For instance, for loose-packed lattices such as the body-centred cubic, all graphs which contain polygons with an odd number of sides e.g. triangles, pentagons, tetrahedra etc. cannot occur and their lattice constants are zero. The fundamental parameters that characterize any given lattice are its coordination number ' q ' and the lattice constants of the closed connected graphs i.e. connected graphs whose vertices are all of degree greater than or equal to two. For the well known lattices, many of these constants are listed by Domb (1960) in his major review article on critical phenomena. All other lattice constants namely of disconnected and/or open graphs can be expressed in terms of these parameters. The techniques used are very clearly described by Domb (1960) and he also lists many of these additional constants. It is important to note that the lattice constant for a connected graph is directly proportional to N , the number of sites in a given regular lattice. For a disconnected graph, however, con-

sisting of n connected components, the lattice constant has terms proportional to $N^n, N^{n-1} \dots N$. As we mentioned earlier, in transforming from Z to $\log Z$, all terms involving powers of N higher than the first cancel so that for most purposes we are primarily interested only in that part of the lattice constant linear in N .

The most difficult step by far in this method is to enumerate and calculate the traces of all the different products of spin variables associated with the same graph. The fundamental problem is simply that the permutation operators that constitute our interaction do not commute e.g.

$$[P_{12}, P_{23}] \neq 0 \quad (4.10)$$

or equivalently,

$$[a_i^m, a_i^n] \neq 0 \text{ for all } m \neq n. \quad (4.11)$$

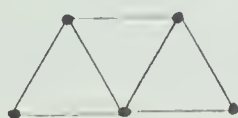
Because of this, for example, the terms $(P_{12} - 1/3)(P_{23} - 1/3)(P_{31} - 1/3)$ and $(P_{12} - 1/3)(P_{31} - 1/3)(P_{23} - 1/3)$ both associated with the graph



have different traces. The number of distinct arrangements

of the interactions all corresponding to the same graph that have different traces increases enormously as the number of bonds in the graph increases. For instance, Rushbrooke and Wood (1958) point out that

graphs such as



give rise to as many as 360

separate terms. The work then is very tedious and lengthy and it is a triumph of ingenuity and perseverance that Rushbrooke and Wood (1958) obtained by this method six terms of the specific heat series for the general spin Heisenberg model.

We should note that this great difficulty is of course absent in such classical models as the Ising model. As a result, Hunter (1967) has calculated twelve terms of the specific heat series for the Ising model on the face-centred cubic lattice by the direct method.

Thus far our remarks have centred round the calculation of the partition function. For the initial susceptibility given by (3.9) and (3.10), we have to calculate χ_r'' . Now from (3.12), μ_r'' is given by,

$$\mu_r'' = \frac{2rC_2}{3^N} \text{trace}(P^{r-2}Q^2) , \quad (4.12)$$

since P and Q commute. As with the calculation of $\text{trace}(P^n)$ so also with (4.12) we have a correspondence between terms in the expansion of the product and linear graphs. Only now our graphs are decorated by the addition of two crosses. These crosses occur on the sites occupied by the S_i^Z terms from Q^2 . Rushbrooke and Wood (1958) discuss three theorems which enable them to calculate the susceptibility series without evaluating any further traces than were required for the partition function series. They were thus able to determine seven terms of the susceptibility series.

B. The Cluster Expansion Method

It seems that the work of Rushbrooke and Wood represents the maximum amount of information that can be obtained practically by this method about quantum mechanical models where the interactions do not commute. It was with this background that Domb (1960) first tentatively suggested the cluster expansion approach (which is an expansion in terms of connected graphs or clusters only). Together with the application of group theory it represents a major advance in technique and enabled Domb and Wood (1965) to obtain for the spin $1/2$ Heisenberg model only, an additional three terms in the susceptibility series for loose packed lattices and two terms for the specific heat series. Subsequently, Rushbrooke et al. (1966) obtained a total of eleven terms in the susceptibility series and nine terms in the specific heat series for loose packed lattices and ten terms and eight terms respectively for the face-centred cubic lattice.

It is also the approach that we have used in our calculation on the spin one exchange model. It has enabled us to calculate more terms and consequently make more accurate predictions regarding critical behavior than has previously been possible for any other spin one model, particularly the spin one Heisenberg model.

At least two detailed and rigorous derivations of the cluster expansion method exist in the literature (Rushbrooke (1964) and Sykes et al. (1966)). Rather than reproduce one or both of these derivations, we

shall try to illustrate as clearly as possible how the cluster expansion can be considered merely as a rearrangement of the direct method expansion.

In the latter, we have just found that the coefficients of the zero-field 'log Z' expansion for a regular lattice L, are expressed in the form,





$$\lambda_r = N \sum_{g_r} [\text{coeff. of } N \text{ in } (g_r; L)] W(g_r) , \quad (4.13)$$

where the summation is over all graphs of r edges including disconnected graphs and multiline graphs i.e. graphs with more than one distinct edge between at least one pair of vertices. For instance, for r=2, three

graphs may contribute namely,  .

The $W(g_r)$ are the weights associated with each graph which in this case are the sums of all the traces of products of spin operators which correspond to each graph.

The first step in the rearrangement is to eliminate effectively any consideration of the disconnected graphs. To do this, we realise that the coefficient of N in the lattice constant of a disconnected graph is the sum of parts each of which can be associated with one of the connected graphs formed from an overlap of the connected components of the disconnected graph. An overlap of a number of graphs in this context means the identification of at least one of the vertices from each graph. To illustrate, the only two overlaps of the disconnected


graph  are  and  . The lattice constant of  on a regular lattice L with coordination number q is given by,

$$\left(\text{graph}; L \right) = \frac{Nq}{4} \left(\frac{Nq}{2} - 2q + 1 \right) . \quad (4.14)$$

The coefficient of N in (4.14) can be split in two parts,

$$a) \quad - \frac{Nq}{2} (q - 1) = - \left(\text{triangle graph}; L \right) , \quad (4.15)$$

$$\text{and} \quad b) \quad - \frac{Nq}{4} = - \frac{1}{2} \left(\text{loop graph}; L \right) . \quad (4.16)$$

In (4.13) therefore, we need not explicitly consider  but instead alter $W(\text{triangle graph})$ by the contribution $-W(\text{two parallel edges})$ and $W(\text{loop graph})$ by the amount $-1/2 W(\text{two parallel edges})$. In other words, (4.13) can be rewritten in the form,

$$\lambda_r = \sum_{g_r} (g_r; L) W^1(g_r) , \quad (4.17)$$

where now the primed summation is over only connected graphs and the weights of the disconnected graphs have been appropriately distributed among the connected graphs. For instance in (4.17), we would find

$$a) \quad W'(\text{triangle}) = W(\text{triangle}) - W(\text{two separate edges}) \quad (4.18)$$






$$b) \quad W'(\text{loop}) = W(\text{loop}) - \frac{1}{2} W(\text{two separate edges}) \quad (4.19)$$

independently of the lattice L .

Thus (3.14) can be written as,

$$\log Z(T) = \sum_{r=0}^{\infty} \sum_{g_r} \frac{K^r}{r!} (g_r; L) W'(g_r), \quad (4.20)$$

where, to repeat, only connected graphs are considered explicitly. The final step in the rearrangement is to group together all terms in (4.20) associated with graphs of the same basic type. Such graphs have the same number of vertices and the same topology and differ only in the multiplicity of their bonds. The basic graph of such a collection is the

graph with no multiple bonds. For instance,   
 etc. are graphs of the same basic type with  being the

basic graph. To see that this regrouping is so vital, we introduce the

concept of labelled lattice constant $(g;G)^*$ which represents the number of weak embeddings of a labelled graph g on a graph G whose sites are themselves distinguishable. Here embeddings are counted separately which differ not in the particular sites of G which are identified with the vertices of g but only in the particular order in which the identification is made. An important relation is that

$$(g;G) = \frac{(g;G)^*}{(g;g)^*} \quad (4.21)$$

where $(g;g)^*$ is often known as the symmetry number of g . For example,

$$\left(\begin{array}{c} \bullet \\ \diagup \quad \diagdown \\ \bullet \quad \bullet \end{array} ; \begin{array}{c} \bullet \\ \diagup \quad \diagdown \\ \bullet \quad \bullet \end{array} \right)^* = 6 \quad (4.22)$$

and

$$\left(\begin{array}{c} \bullet \\ \diagup \quad \diagdown \\ \bullet \quad \bullet \end{array} ; \begin{array}{c} \bullet \\ \diagup \quad \diagdown \\ \bullet \quad \bullet \end{array} \right)^* = 2 \quad (4.23)$$

Let C_i , $i=0,1,2,3,\dots$, denote the connected basic graphs or clusters as we shall now call them. For our purposes it will prove convenient to label the clusters in order of increasing number of bonds, then increasing number of vertices for a given number of bonds and finally by some arbitrary convention for equal numbers of bonds and vertices. If g_r is a graph belonging to the same basic type as C_i , then we have the important identity,

$$(g_r;L)^* = (C_i;L)^* \quad (4.24)$$

From (4.21) we can therefore write,

$$(g_r; L) = \frac{(C_i; L)^*}{(g_r; g_r)^*} \quad (4.25)$$

$$= \frac{(C_i; C_i)^* (C_i; L)^*}{(g_r; g_r)^* (C_i; C_i)^*} . \quad (4.26)$$

Again using (4.21) we obtain from (4.25)

$$(g_r; L) = \frac{(C_i; C_i)^*}{(g_r; g_r)^*} (C_i; L) . \quad (4.27)$$

Substitution of (4.27) in (4.20) then yields,

$$\log Z(T) = \sum_i (C_i; L) \phi_i(K) \quad (4.28)$$

$$\phi_i(K) = (C_i; C_i)^* \sum_{g_r} \frac{K^r}{r!} \frac{W^r(g_r)}{(g_r; g_r)^*} \quad (4.29)$$

In (4.28) the summation is over connected clusters only and in (4.29) it is over all graphs of the same basic type as C_i .

(4.28) constitutes the basic relation in the cluster expansion method. The outstanding feature of this method is that the $\phi_i(K)$ are

not found by considering (4.29) which would be just as difficult and laborious as the direct method itself. Rather we realise that (4.28) can be applied to any finite system as well as the regular lattice L . This means we can write

$$\log Z(C_i, K) = \sum_{j=0}^i (C_j; C_i) \phi_j(K) . \quad (4.30)$$

The $\phi_i(K)$ are obtained by successively solving (4.30) for increasing i or in other words, they are found recursively from the relation,

$$\phi_{i+1}(K) = \log Z(C_{i+1}; K) - \sum_{j=0}^i (C_j; C_{i+1}) \phi_j(K) . \quad (4.31)$$

It is important to note that the $\phi_i(K)$ depend only on C_i and not on the system to which the cluster expansion (4.28) is being applied. Thus the $\phi_i(K)$ obtained from (4.30) and (4.31) are the same $\phi_i(K)$ we substitute in (4.28) to obtain $\log Z$ for any regular lattice. The only feature in (4.28) that characterizes the particular lattice under consideration is the lattice constants $(C_i; L)$.

To illustrate the procedure, let us consider $i=0 \rightarrow 3$. (4.30)

gives

$$\log Z (\bullet, K) = \phi_0(K) ,$$

$$\log Z (\bullet \text{---} \bullet, K) = 2\phi_0(K) + \phi_1(K) ,$$

(4.32)

$$\log Z (\bullet \begin{array}{c} \nearrow \\ \searrow \end{array} \bullet, K) = 3\phi_0(K) + 2\phi_1(K) + \phi_2(K) ,$$

$$\log Z (\triangle, K) = 3\phi_0(K) + 3\phi_1(K) + 3\phi_2(K) + \phi_3(K) .$$

The left-hand side of (4.32) is evaluated in each case as a series in K of the form (3.14). Since these are the partition functions of finite clusters, their calculation presents no problem in principle and can be performed on a computer. For instance for $C_3 = \triangle$ the Hamiltonian is given by,

$$\mathcal{H} = -J(P_{12} + P_{23} + P_{31} - 1) , \quad (4.33)$$

whose matrix representation has dimensions 27×27 . To obtain μ_r , we merely have to evaluate trace (\mathcal{H}^r) and then use the cumulant-moment identities in appendix A to obtain $\log Z(\triangle, K)$ as a series in K . We then solve (4.32) for $\phi_i(K)$, $i=0 \rightarrow 3$, and to obtain their contribution to $\log Z(K)$ for any given regular lattice we substitute in (4.28) using the appropriate lattice constants.

A completely analogous procedure can be followed to obtain the initial magnetic susceptibility. For instance, corresponding to (4.28)

and (4.29), we would have,

$$\bar{\chi}_0(T) = \sum_i (C_i; L) \psi_i(K) \quad (4.34)$$

$$\psi_i(K) = (C_i; C_i)^* \sum_{g_r} \frac{K^r}{(r+2)!} \frac{V'(g_r)}{(g_r; g_r)^*} \quad (4.35)$$

From (3.18) and (4.12) we see that the graphs g_r in (4.35) have r edges with two of the vertices decorated with crosses.

Finally, an important feature of both the $\phi_i(K)$ and $\psi_i(K)$ is that the lowest power of K in their series that can have a non-zero coefficient is K^b with b equal to the number of bonds in C_i . This is easily understood since in both (4.29) and (4.35) the summation is over all graphs of the same basic type as C_i . The graph with the smallest number of bonds in this collection is, of course, C_i itself which contributes to that power of K which equals its number of bonds. This means then that to obtain the exact coefficients in (4.28) and (4.34) for any infinite lattice up to K^b , we have to determine at most the series for $\phi_i(K)$ and $\psi_i(K)$ up to K^b corresponding to all graphs with a maximum of b bonds.

In summary, while the direct method involves consideration of both connected, disconnected and multiline graphs, the cluster expansion is concerned only with connected basic graphs. Whereas the calculation of weights in the direct method involves an enumeration of many different

traces corresponding to any given graph, in the cluster expansion method the weights $\phi_i(K)$ and $\psi_i(K)$ present no such difficulties and can be calculated by computer. The advantages then are obvious.

The only practical difficulty arises in the calculation of the traces for the larger finite clusters. In our calculation, for instance, to obtain the coefficient of K^7 , it is necessary to consider clusters with up to seven vertices. The corresponding matrices are 2187 x 2187! It is quite impossible for the computing facilities available to us to manipulate such large matrices. However, since our interaction Hamiltonian consists of a sum of permutation operators, we were able to make use of the powerful formalism of group theory applied to the particular case of the symmetric group. This essentially enabled us to perform a similarity transformation on our matrix representation so that our matrices are 'block diagonalised' or 'reduced'. As an indication of the success of this approach, for the 2187 x 2187 matrices the largest matrix we in fact considered was 35 x 35.

In the next chapter, we shall describe how group theory has been applied to this problem both to determine $\log Z$ and χ .

CHAPTER V

APPLICATION OF GROUP THEORYA. Concepts in Group Theory

As a preliminary we shall define a number of the terms in group theory that we shall use frequently in this chapter. A much more complete account of these basic concepts can be found in many books on the subject of which Wigner's (1959) is perhaps the best known.

A group is a set of objects (the elements of the group) among which a law of combination is defined having the following characteristics.

a) It satisfies the closure property; namely, if A and B are elements of the group and under the law of combination we obtain

$$AB = C , \quad (5.1)$$

then C must also be an element of the group.

b) The associative law must hold i.e.

$$(AB)C = A(BC) . \quad (5.2)$$

c) There exists among the elements of the group an identity element E such that

$$XE = EX = X \quad (5.3)$$

where X is any element of the group.

d) To every element X of the group there corresponds a reciprocal element X^{-1} of the group such that

$$XX^{-1} = X^{-1}X = E \quad . \quad (5.4)$$

The above four conditions are often known as the group axioms or postulates.

The number of elements in a group is known as the order of the group.

If A and B are two elements of a group such that

$$B = X^{-1}AX \quad (5.5)$$

where X is some element of the group, then B is said to be conjugate to A . We define the class of A to be the collection of elements $X^{-1}AX$ where X runs over all the elements of the group i.e. the class of A is all those elements that are conjugate to A .

A set of elements of a group which itself forms a group is called a subgroup.

Two groups G and G' are said to be isomorphic if the elements of G are in one-to-one correspondence with the elements of G' such that, if X and Y of G correspond to X' and Y' of G' , then XY of G corresponds to $X'Y'$ of G' . On the other hand G is said to be homomorphic to G' if each

element of G' corresponds to at least one element of G such that if X and Y of G correspond to X' and Y' of G' then XY of G corresponds to $X'Y'$ of G' . Evidently while isomorphism is a reciprocal property, homomorphism is not.

A representation of a group G is a matrix group onto which the group to be represented is homomorphic i.e. to every element X of G there corresponds a matrix $D(X)$ such that

$$D(X)D(Y) = D(XY) \quad (5.6)$$

where the law of combination of the matrices is ordinary matrix multiplication. It is clear that these matrices must be square and the number of rows or columns is known as the dimension of the representation. If the matrix group is isomorphic to the group it represents, then the representation is said to be faithful. Two representations which are related by a similarity transformation are said to be equivalent representations. Such representations have identical traces for corresponding matrices and in the formalism of group theory this means there is no essential difference between them.

From two representations $D(X)$ and $D'(X)$ which may or may not be equivalent it is possible to form a third representation $D''(X)$ given by,

$$D''(X) = \begin{pmatrix} D(X) & 0 \\ 0 & D'(X) \end{pmatrix}, \quad (5.7)$$

where as before X represents any element of the group. $D''(X)$ and any equivalent representation are said to form reducible representations of the group. In the form (5.7) we say $D''(X)$ is a direct sum of $D(X)$ and $D'(X)$. Representations for which a transformation to the form (5.7) is not possible are said to be irreducible. If $D(X)$ and $D'(X)$ are, however, irreducible representations then $D''(X)$ alone is said to be in completely reduced form. This concept can be naturally extended to representations of the form (5.7) but containing more than two irreducible representations along the principal diagonal.

If X and Y are two elements of a group G belonging to the same class then it is an easy matter to show that

$$\text{trace } D(X) = \text{trace } D(Y) \quad (5.8)$$

where $D(X)$ is a representation of G . The traces of the matrices corresponding to a set of elements of G chosen one from each class is known as the character table of the representation. Evidently equivalent representations have the same character table. Indeed for most purposes in group theory all one needs to know about a representation is its character table.

One of the important features of the formalism is that associated with any group there exists a finite number of inequivalent irreducible representations usually specified by their character tables. The main task for which the formalism is used in its applications is to reduce

to its completely reduced form any given reducible representation for which perhaps only the character table is known. How many times each irreducible representation occurs in the completely reduced form is determined by the following important result from the representation theory of groups (e.g. Wigner 1959):

$$n(\gamma) = \frac{1}{g} \sum_R \text{tr} \chi(R) \text{tr} \Gamma(R) \quad (5.9)$$

where γ labels the irreducible representations and Γ the reducible representation, $n(\gamma)$ is the number of times the γ irreducible representation occurs, g is the order of the group, $\text{tr} \chi(R)$ is the trace of the matrix in the representation γ corresponding to the element R and the summation is over all the elements of the group. It should be noted that in (5.9) the $n(\gamma)$ are obtained only from a knowledge of the character tables of the representations.

B. Application to the Symmetric Group

Our task in this section is two-fold. Firstly we wish to determine the completely reduced form of a particular reducible representation of the symmetric group which means essentially using (5.9). However, for our purposes a knowledge of only the number of times a given irreducible representation occurs is not sufficient. We require in addition to determine the explicit form of the matrices in each irreducible rep-

resentation that occurs. Then the determination of the coefficients in the finite cluster expansions for $\log Z$ and χ presents few difficulties.

Before discussing both these steps in detail, we shall introduce some of the relevant features of the symmetric group. The elements of the symmetric group Π_N of the N th degree are the permutations or the interchanges of N objects. The order of the group is then $N!$ A Permutation which replaces 1 by α_1 , 2 by α_2 ..., and finally N by α_N is denoted by $\begin{pmatrix} 1 & 2 & 3 & \dots & N \\ \alpha_1 & \alpha_2 & \alpha_2 & \dots & \alpha_N \end{pmatrix}$. An interesting property of all $N!$ permutations is that they can be resolved into a product of cycles. A cycle $(r_1 r_2 \dots r_\lambda)$ is a permutation which replaces every element r_k by the element following it r_{k+1} , except the last element r_λ which is replaced by the first element r_1 . The cycle $(r_1 r_2 \dots r_\lambda)$ is then identical with the permutation $\begin{pmatrix} r_1 & r_2 & \dots & r_\lambda \\ r_2 & r_3 & \dots & r_1 \end{pmatrix}$. As an example the permutation $\begin{pmatrix} 1 & 2 & 3 & 4 & 5 & 6 & 7 \\ 3 & 4 & 5 & 6 & 1 & 7 & 2 \end{pmatrix}$ can be rearranged by interchanging columns (which of course does not alter the permutation) to give $\begin{pmatrix} 1 & 3 & 5 & 2 & 4 & 6 & 7 \\ 3 & 5 & 1 & 4 & 6 & 7 & 2 \end{pmatrix}$ from which it is obvious that the decomposition into cycles is

$$\begin{pmatrix} 1 & 2 & 3 & 4 & 5 & 6 & 7 \\ 3 & 4 & 5 & 6 & 1 & 7 & 2 \end{pmatrix} = (1 \ 3 \ 5)(2 \ 4 \ 6 \ 7) . \quad (5.10)$$

The cycle structure of this permutation is denoted by $(3,4)$ the numbers denoting the length of each cycle in the permutation. To generalise, the

cycle structure of a permutation from Π_N is given by a partition of the positive integer N into an ordered set of positive integer whose sum is N . Typically the partition $(1^{\beta_1}, 2^{\beta_2}, 3^{\beta_3}, \dots, N^{\beta_N})$, where we have

$$\sum_{r=1}^N r\beta_r = N, \quad (5.11)$$

represents a permutation containing β_1 cycles involving one object, β_2 cycles involving two objects, ..., and finally β_N cycles involving N objects.

One can see almost by inspection that permutations with the same cycle structure belong to the same class of the symmetric group. Indeed, the number of classes in Π_N is equal to the number of partitions of N (the order of the integers in the partition is of no consequence) and we label each class by its corresponding partition. In particular the interaction (2.18) with which we are primarily concerned is the transposition or simple permutation involving only two objects and all such simple permutations belong to the class of Π_N denoted by $(1^{N-2}, 2)$.

One of the well known results of the general representation theory of groups is that the number of irreducible representations associated with a group is equal to the number of its classes. In the case of the symmetric group Π_N , it is also possible to set up a one-to-one correspondence between the irreducible representations and the partitions of N (e.g. Wigner 1959). We then also label the irreducible representa-

tions by the partitions, only now we write the numbers in descending order to avoid ambiguity. The character tables of all the irreducible representations of Π_N for $N = 2, 3 \dots 7$ are reproduced from Littlewood (1940) in appendix C.

In the case of a cluster of N vertices, the reducible representation (of dimension 3^N) that we consider is the direct product representation in which the space of the N spin one systems is constructed as the direct product of N single spin spaces. It is then possible to determine almost by inspection the character table of the representation. As an example of the simple procedure used we shall consider the case of $N=3$ in detail. We represent the three linearly independent single spin one states by α, β, γ . The $3^3 = 27$ basic states of the direct product representation are given by $\delta\epsilon\zeta$ where $\delta\epsilon\zeta = \alpha, \beta$ or γ . Since there are nine states of the form $\delta\delta\epsilon$, the trace of the simple transposition operator which interchanges the first two symbols and hence all members of the class (12) is nine. Likewise there are three states of the form $\delta\delta\delta$, so that the trace of the operator that permutes all three symbols i.e. the class (3) is three. The character table of our reducible representation of Π_3 is given by ,

(1^3)	(12)	(3)	
27	9	3	(5.12)

In similar fashion the character table of the direct product reducible

representation of Π_N for $N \leq 7$ can easily be obtained and the results are given in appendix C.

We are now in a position to apply (5.9) to determine $n(\gamma)$ for each irreducible representation of Π_N for $N \leq 7$. The calculation is straightforward and the results obtained are reproduced in appendix C. To obtain the series for $\log Z$ for any given finite cluster of N vertices we can write

$$\mu_n = \text{trace} (P^n) \quad (5.13)$$

$$= \sum_{\gamma} n(\gamma) \text{ trace} (P_{\gamma}^n) \quad (5.14)$$

where P_{γ} is the matrix representing P in the irreducible representation γ of Π_N . Then using the cumulant-moment identities given in appendix A, we immediately obtain $\log Z$ in the form (3.14). The great simplification introduced in the transition from (5.13) to (5.14) is worth emphasizing. For $N = 7$ for instance, the dimension of the matrices in (5.13) is $2187 = 3^7$ whereas the largest matrix in (5.14) has a dimension of 35.

There remains however, the task of determining the explicit form of the permutation matrices (i.e. of those matrices representing the class $(1^{N-2}, 2)$ of Π_N) in any irreducible representation. The technique we use is based largely on a method first introduced by Yamanouchi (1937). It is essentially an inductive procedure in which, in order to calculate the irreducible representations of Π_N we assume a complete

knowledge of the irreducible representations of both Π_{N-1} and Π_{N-2} .

Let us suppose that we wish to determine the details of the irreducible representation D of Π_N associated with the partition (f_1, f_2, \dots, f_k) where

$$\text{a) } f_1 \geq f_2 \geq f_3 \geq \dots \geq f_k \geq 0 , \quad (5.15)$$

$$\text{b) } \sum_{r=1}^k f_r = N .$$

The matrices of D which do not contain the index N i.e. belong to the subgroup Π_{N-1} of Π_N are formed by the direct sum of the corresponding matrices in the irreducible representations of Π_{N-1} associated with the partitions,

$$(f_1-1, f_2, \dots, f_k) ,$$

$$(f_1, f_2-1, \dots, f_k) ,$$

$$(f_1, f_2, f_3-1, \dots, f_k) , \quad (5.16)$$

- - - - -
- - - - -

$$(f_1, f_2, \dots, f_{k-1}),$$

excepting these partitions in which the f 's are not in decreasing order.

The irreducible representation associated with $(f_1, f_2, \dots, f_{r-1}, \dots, f_k)$ will be denoted by D_r . We shall use the suffix 'r' to label the rows and columns of D in which the matrices D_r occur.

If we consider only the elements belonging to the subgroup Π_{N-2} , then D_r itself will take the form of a direct sum of the irreducible representations D_{rs} of Π_{N-2} where D_{rs} corresponds to the partition $(f_1, f_2, \dots, f_{r-1}, \dots, f_{s-1}, \dots, f_k)$. The sum is for fixed r and s varying from 1 to k where, as before, cases in which the f 's are not in decreasing order after subtraction are omitted.

These irreducible representations fall into three categories,

- a) D_{rr} (only if $f_r \geq f_{r+1} + 2$) occurs once,
- b) $D_{rs} = D_{sr}$ arise twice if both do not violate the decreasing order of the integers in the partitions,
- c) D_{rs} is possible but D_{sr} is not e.g. if $s = r - 1$ and $f_{r-1} = f_r$.

The rows and columns within the group previously labelled r are now subdivided and denoted by the suffix rs . The rs rows and columns indicate at which point on the principal diagonal the matrix associated with D_{rs} will be placed.

The problem remains of calculating the permutation matrices involving the symbol N . However, it is sufficient to determine only $P_{N-1, N}$ since

$$P_{i, N} = P_{N-1, N} P_{i, N-1} P_{N-1, N} \quad (5.17)$$

(for all $i \neq N$)

and we have just described how the $P_{i,N-1}$ are obtained.

Since $P_{N-1,N}$ commutes with all the elements of Π_{N-2} , we make use of another of the important theorems of the representation theory of groups known as Schur's lemma which states:

If we consider two irreducible representations $D^1(X)$ and $D^2(X)$ of the same group, of dimension ℓ_1 and ℓ_2 respectively where X is a typical element in that group and there exists a matrix M with ℓ_2 rows and ℓ_1 columns such that

$$MD^1(X) = D^2(X)M \quad (5.18)$$

for all X , then we conclude either

- 1) $M = 0$, provided $\ell_1 \neq \ell_2$,
- 2) $M = 0$ if $D^1(X)$ and $D^2(X)$ are not equivalent representations with $\ell_1 = \ell_2$,
- 3) if $D^1(X)$ and $D^2(X)$ are equivalent representations and $\ell_1 = \ell_2 = \ell$, M is a non-singular ℓ dimensional square matrix, or
- 4) if $D^1(X)$ and $D^2(X)$ are the same representation, M is given by,

$$M = \lambda I \quad , \quad (5.19)$$

where λ is a constant and I is the ℓ dimensional unit matrix.

If now we subdivide the rows and columns of $P_{N-1,N}$ and label them 'rs' just as we did for the elements of Π_{N-2} , we obtain, since $P_{N-1,N}$ commutes with Π_{N-2} ,

$$V_{rs,rs} P_{rs,pq} = P_{rs,pq} V_{pq,pq} \quad (5.19)$$

where $V_{rs,pq} = V_{rs,rs} \delta_{rs,pq}$ is the representation matrix for any element of Π_{N-2} in the irreducible representation D_{rs} and $P_{rs,pq}$ is the block in the subdivided form of $P_{N-1,N}$ occurring at the intersection of rows rs and columns pq.

According to Schur's lemma, we must have that

$$P_{rs,pq} = 0 \quad (5.20)$$

unless $pq = rs$ or sr in which case

$$P_{rs,rs} = \lambda_{rs,rs} I_{rs,rs} \quad (5.21)$$

and

$$P_{rs,sr} = \lambda_{rs,sr} I_{rs,sr} \quad (5.22)$$

where $I_{a,b}$ is the unit matrix occurring at the intersection of rows a and columns b. Since $P_{N-1,N}^2 = I$, we have for indices corresponding to the types a) and c) discussed above that

$$\left| \lambda_{rs,rs} \right|^2 = 1 , \quad (5.23)$$

and for type b),

$$\left| \lambda_{rs,rs} \right|^2 + \left| \lambda_{rs,sr} \right|^2 = 1 . \quad (5.24)$$

Thus we require only the diagonal elements $\lambda_{rs,rs}$ to construct $P_{N-1,N}$. To do this, we first label by P_{ij} the matrix representing the transposition of the i th and j th symbols in the irreducible representation D . It can easily be shown that $\sum_{i < j}^N P_{ij}$ i.e. the sum of all the elements of the class $(1^{N-2}, 2)$ commutes with all members of Π_N and therefore by Schur's lemma we can write

$$\sum_{i < j}^N P_{ij} = cI \quad (5.25)$$

where c is some constant. Taking traces of both sides we immediately obtain the identity,

$$\frac{N(N-1)}{2} T = cd , \quad (5.26)$$

where d is the dimension of D and T is the trace of any of the matrices belonging to the class $(1^{N-2}, 2)$ in the representation D (given in appendix C). c is then given by,

$$c = \frac{N(N-1)T}{2d} . \quad (5.27)$$

Similarly in the irreducible representation D_r , we can write

$$\sum_{i < j=1}^{N-1} P_{ij}^{(r)} = c_r I_r, \quad (5.28)$$

and in the irreducible representation D_{rs} ,

$$\sum_{i < j=1}^{N-2} P_{ij}^{(rs)} = c_{rs} I_{rs}, \quad (5.29)$$

where I_r and I_{rs} are unit matrices of dimension d_r and d_{rs} respectively.

Corresponding to (5.27) we have

$$c_r = \frac{(N-1)(N-2)T_r}{2d_r}; \quad c_{rs} = \frac{(N-2)(N-3)}{2d_{rs}} T_{rs},$$

where T_r and T_{rs} are the traces of the permutation operators in the representations D_r and D_{rs} .

It is clear from (5.25) and (5.28) that the matrix A_N given by,

$$A_N = \sum_{i=1}^{N-1} P_{i,N} = \sum_{i < j=1}^N P_{ij} - \sum_{i < j=1}^{N-1} P_{ij}, \quad (5.31)$$

is diagonal with sub-matrices $(c - c_r)I_{r,r}$ along the principal diagonal.

Similarly from (5.28) and (5.29) we can see that A_{N-1} defined by,

$$A_{N-1} = \sum_{i=1}^{N-2} P_{i,N-1} \quad , \quad (5.32)$$

is a diagonal matrix with sub-matrices $(c_r - c_{rs})I_{rs,rs}$ occurring on the principal diagonal.

From (5.31) we can write immediately that

$$A_N P_{N-1,N} = \sum_{i=1}^{N-2} P_{i,N} P_{N-1,N} + I \quad (5.33)$$

and, multiplying both sides by $(P_{N-1,N})^2 = I$, this becomes

$$P_{N-1,N} \sum_{i=1}^{N-2} P_{i,N-1} + I = A_N P_{N-1,N} \quad , \quad (5.34)$$

i.e.

$$A_N P_{N-1,N} - P_{N-1,N} A_{N-1} = I \quad . \quad (5.35)$$

Writing (5.35) in terms of the submatrices labelled rs , pq for the two cases $pq = rs$ and $pq = sr$, we obtain

$$(c - c_r)P_{rs,rs} - P_{rs,rs}(c_r - c_{rs}) = I_{rs,rs} \quad , \quad (5.36)$$

and
$$(c - c_r)P_{rs, sr} - P_{rs, sr}(c_s - c_{sr}) = 0 \quad (5.37)$$

where $c_{rs} = c_{sr}$. Substituting (5.21) and (5.22) in (5.36) and (5.37) we get

$$\lambda_{rs, rs} = \frac{1}{c - 2c_r + c_{rs}} \quad (5.38)$$

and $\lambda_{rs, sr} = 0$ unless $c - c_r - c_s + c_{rs} = 0$. (5.38) together with (5.24) are the equations we use to determine $P_{N-1, N}$ and complete our calculation of the irreducible representation D. An arbitrariness in sign that will arise in the solution of (5.24) just indicates that this method may yield several equivalent representations.

We should add that in applying this technique we assume a knowledge of the irreducible representations of π_2 and π_3 . They are quite trivial, however, and can be determined almost by inspection. As an example of this procedure, we shall consider the six-dimensional irreducible representation $(3, 1^2)$ of π_5 corresponding to the partition $(3, 1, 1)$. In (5.15) then we have

$$f_1 = 3; f_2 = 1; f_3 = 1. \quad (5.39)$$

The matrices of this representation belonging to the subgroup π_4 are the direct sum of the irreducible representations $(2, 1^2)$ and $(3, 1)$ of π_4 , labelled according to our procedure as D_1 and D_3 respectively i.e. of the form,

$$\begin{pmatrix} (2,1^2) & 0 \\ 0 & (3,1) \end{pmatrix}, \quad (5.40)$$

where both submatrices are 3-dimensional. Similarly the matrices of the subgroup Π_3 are of the form,

$$\begin{array}{cccc} & 11 & 13 & 31 & 32 \\ \begin{array}{c} 11 \\ 13 \\ 31 \\ 32 \end{array} & \begin{pmatrix} (1^3) & 0 & 0 & 0 \\ 0 & (2,1) & 0 & 0 \\ 0 & 0 & (2,1) & 0 \\ 0 & 0 & 0 & (3) \end{pmatrix} & & & \end{array} \quad (5.41)$$

If the irreducible representations forming the submatrices of (5.14), $D_{11} \equiv (1^3)$ is 1-dimensional and of the type a), $D_{13} \equiv D_{31} \equiv (2,1)$ is 2-dimensional and of type b) and $D_{32} \equiv (3)$ is 1-dimensional and of type c). Also in (5.41) we have subdivided the rows and columns of the matrix and labelled them according to our procedure outlined above.

To determine P_{45} , we use (5.27) and (5.30) together with the character tables in appendix C to find that

$$\left. \begin{array}{l} c = 0, \\ c_1 = -2; \quad c_3 = +2; \\ c_{11} = -3; \quad c_{13} = 0 = c_{31}; \quad c_{32} = +3. \end{array} \right\} \quad (5.42)$$

From (5.38) we then get

$$\lambda_{11,11} = +1; \quad \lambda_{13,13} = \frac{1}{4}; \quad \lambda_{31,31} = -\frac{1}{4}, \quad (5.43)$$

and finally using (5.24) that

$$\lambda_{13,31} = \pm \frac{\sqrt{15}}{4} = \lambda_{31,13}. \quad (5.44)$$

A possible form for P_{45} in the irreducible representation $(3,1^2)$ of π_5 is therefore given by,

$$P_{45} = \begin{array}{c} \begin{array}{cc} & \begin{array}{cc} 11 & 13 \end{array} \\ \begin{array}{c} 11 \\ 13 \\ 31 \\ 32 \end{array} & \begin{pmatrix} \begin{array}{cc|cc|c} +1 & 0 & 0 & 0 & 0 & 0 \\ 0 & \frac{1}{4} & 0 & -\frac{\sqrt{15}}{4} & 0 & 0 \\ 0 & 0 & \frac{1}{4} & 0 & -\frac{\sqrt{15}}{4} & 0 \\ 0 & -\frac{\sqrt{15}}{4} & 0 & -\frac{1}{4} & 0 & 0 \\ 0 & 0 & -\frac{\sqrt{15}}{4} & 0 & -\frac{1}{4} & 0 \\ 0 & 0 & 0 & 0 & 0 & -1 \end{array} \end{pmatrix} \end{array} \quad (5.45)$$

An exactly similar procedure was followed for all the irreducible representations γ for which $n(\gamma)$ given in appendix C was non-zero. In fact, the whole calculation of finding the explicit form of the irreducible representations, substituting in (5.14) and then finding the cumulants was performed in one computer programme.

Thus far our discussion has been aimed at deriving the $\log Z$ series expansions. However, in determining the initial susceptibility series, no further traces have to be calculated. Only a different linear combination from (5.14) is required to find μ_r'' given by (4.12). To make this point clear, it is convenient to visualise the transformation from the direct product representation to the completely reduced representation in two stages. The intermediate stage is a transformation to a representation in which the basic states are simultaneous eigenstates of $S^2 = (S_1 + S_2 + \dots + S_N)^2$ and Q . Since

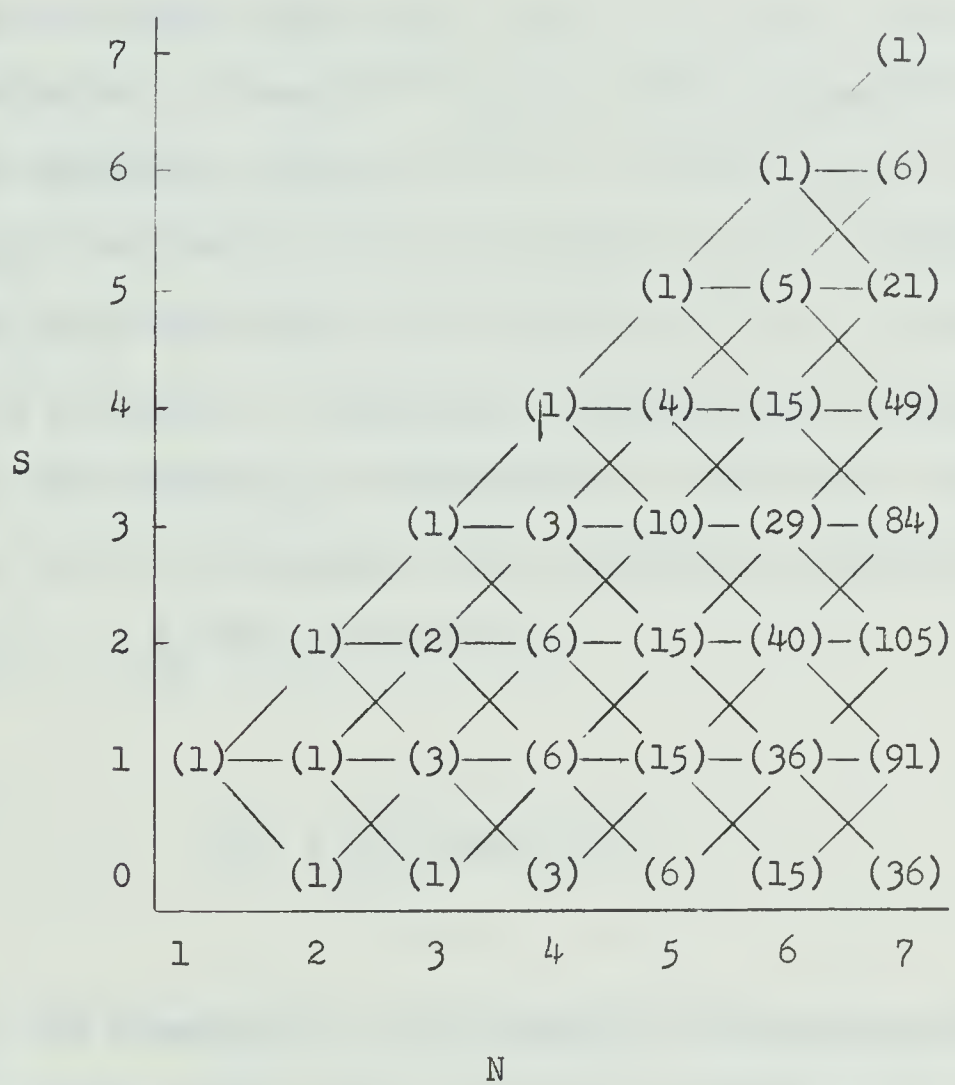
$$[P, S^2] = [P, Q] = [P, S_{\pm}] = 0 \quad (5.46)$$

where $S_{\pm} = (S_1^x + S_2^x + \dots + S_N^x) \pm i(S_1^y + S_2^y + \dots + S_N^y)$, P will have no non-zero matrix elements between states of different S^2 and Q quantum numbers. In addition, for constant S^2 , the non-zero matrix elements will be independent of the Q quantum number. In this representation, P will consist of a direct sum of matrices each associated with a value of the quantum numbers S of S^2 , where $S = N, N-1, N-2, \dots, 1$, or 0 with $P(S)$ being repeated $2S+1$ times. The notation $P(S)$ is introduced here to denote the matrix representing P in the representation in which S^2 and Q are diagonal with S fixed.

The dimensions of $P(S)$ are given in the 'branching' diagram in figure 5.1 for all S and $N \leq 7$. It is constructed according to the well known rules for addition of angular momenta in quantum mechanics. We

Fig. 5.1

Branching diagram for spin one particles. N is the number of particles, S the total spin and the numbers in brackets indicate the degeneracy of the corresponding state for fixed S^Z .



then have in this representation, from (4.12), that

$$\mu_r'' = \frac{2_r C_2}{3^{N+1}} \sum_{S=0}^N \text{trace} \left\{ P^{r-2}(S) \right\} S(S+1)(2S+1) \quad (5.47)$$

where we have already summed (for fixed S) over all Q from $-S$ to $+S$. The determination of $\text{trace} \left\{ P^{r-2}(S) \right\}$ is further simplified by transforming to the completely reduced form. For a given value of N , this actually entails distributing the known numbers of the irreducible representations among the various $P(S)$ for $S = 0, 1, 2, \dots$, and N . This is achieved by a process of elimination satisfying the following constraints.

1) The irreducible representation γ must occur a total of $n(\gamma)$ times i.e. if it is associated in the completely reduced form with $S = S_1, S_2, \dots, S_k$ then we must have

$$n(\gamma) = \sum_{r=1}^k (2S_r + 1) . \quad (5.48)$$

2) The dimensions of the irreducible representations associated with a particular value of S must add up to the dimensions of the $P(S)$ as given in the branching diagram.

3) For fixed N , the one dimensional irreducible representation $n(N)$ will occur once in the reduced form of $P(S)$ for $S = N, N - 2, N - 4, \dots$ etc. We must have therefore

$$n(N) = \sum_{r=0}^{\frac{N}{2}} (2N - 4r + 1) \quad (5.49)$$

where the summation in (5.49) is over all positive integer values of $r \leq \frac{N}{2}$.

This last condition requires some explanation. We shall illustrate for the case of $N = 4$ but the argument is exactly the same for all values of N . To find eigenfunctions of S^2 and Q which might correspond to the irreducible representation (4), we require combinations of products of the $\alpha(i)$, $\beta(i)$, and $\gamma(i)$, the single particle eigenfunctions of S_i^2 , that are symmetric under all the operations of the symmetric group. For $Q = 4$, there is only one possibility namely $\alpha\alpha\alpha\alpha$ where the labelling of the spins has been omitted. This must also be an eigenstate of S^2 and hence (4) must be associated with the $S = 4$, $Q = 4$ eigenstate. Because of the independence of $P(S)$ on Q , we therefore associate nine of the occurrences of (4) with the $S = 4$, $Q = +4 \rightarrow -4$ eigenstates. For $Q = 3$, there is likewise only one possible completely symmetric eigenstate which must also be then a simultaneous eigenstate of S^2 with $S = 4$. It is of the form $\alpha\alpha\alpha\beta + \alpha\alpha\beta\alpha + \alpha\beta\alpha\alpha + \beta\alpha\alpha\alpha$. For $Q = 2$, there are two linearly independent eigenstates namely $\alpha\alpha\alpha\gamma + \alpha\alpha\gamma\alpha + \alpha\gamma\alpha\alpha + \gamma\alpha\alpha\alpha$ and $\alpha\alpha\beta\beta + \beta\beta\alpha\alpha + \alpha\beta\alpha\beta + \beta\alpha\beta\alpha + \alpha\beta\beta\alpha + \beta\alpha\alpha\beta$. Either one of these or a linear combination of both (it does not matter which for our purpose) must be a simultaneous eigenstate of S^2 with $S = 4$ and the other state or the other lin-

early independent combination must be an eigenstate with $S = 2$. We have therefore an additional five occurrences of the irreducible representation (4) associated with $S = 2$. For $Q = 1$, there are again only two possibilities namely $\alpha\alpha\beta\gamma + \alpha\alpha\gamma\beta + \dots$ a total of $\frac{4!}{2!} = 12$ different orderings of the four symbols and $\alpha\beta\beta\beta + \beta\alpha\beta\beta + \beta\beta\alpha\beta + \beta\beta\beta\alpha$. Hence (4) can not be associated with $S = 1$. For $Q = 0$, however, there are three linearly independent eigenstates given by $\beta\beta\beta\beta$, $\alpha\alpha\gamma\gamma + \dots$ a total of $\frac{4!}{2!2!} = 6$ different orderings and $\alpha\beta\beta\gamma + \dots$ a total of $\frac{4!}{2!} = 12$ different orderings. One of these states or one linear combination must also be an eigenstate of S^2 with $S = 0$ and so we have one more occurrence of (4). The total number of occurrences is fifteen and from appendix C we see that this concurs with (5.49) for $N = 4$.

Finally, as an example of how all these constraints are used, we shall consider the case of $N = 5$. From appendix C, we know that

$$\left. \begin{array}{llll} n(5) & = 21 & \text{with dimension} & = 1 , \\ n(4,1) & = 24 & " & " = 4 , \\ n(3,2) & = 15 & " & " = 5 , \\ n(2^2,1) & = 3 & " & " = 5 , \\ n(3,1^2) & = 6 & " & " = 6 , \end{array} \right\} \quad (5.50)$$

and all other $n(\gamma) = 0$. This distribution has to be fitted to the block diagonalisation given in figure 5.1 for $N = 5$ i.e.

$$\begin{array}{rcl}
 \text{for } S = 5, & \text{the dimension of the block is} & 1, \\
 " & S = 4, & " \quad " \quad " \quad " \quad " \quad 4, \\
 " & S = 3, & " \quad " \quad " \quad " \quad " \quad 10, \\
 " & S = 2, & " \quad " \quad " \quad " \quad " \quad 15, \\
 " & S = 1, & " \quad " \quad " \quad " \quad " \quad 15, \\
 " & S = 0, & " \quad " \quad " \quad " \quad " \quad 6.
 \end{array} \quad (5.51)$$

From constraint 3) the representation (5) occurs eleven times in the $S = 5$ block, seven times in the $S = 3$ and three times in the $S = 1$. The total of 21 agrees with (5.50). The only possible location for the $(2^2, 1)$ representation is the $S = 1$ block. Then from (5.48) we must have $(3, 1^2)$ distributed so that five of its occurrences are in the $S = 2$ block and one in the $S = 0$ block. Nine of the $(4, 1)$ must go to the $S = 4$ block and so on. By this procedure, we obtain a unique distribution of the irreducible representations so that all the constraints are satisfied i.e.

$$\begin{array}{rcl}
 \text{for } S = 5, & \text{the irreducible representations are} & (5), \\
 " & S = 4, & " \quad " \quad " \quad (4, 1), \\
 " & S = 3, & " \quad " \quad (3, 2) + (4, 1) + (5), \\
 " & S = 2, & " \quad (3, 1^2) + (3, 2) + (4, 1), \\
 " & S = 1, & " \quad (3, 2) + (2^2, 1) + (4, 1) + (5), \\
 " & S = 0, & " \quad (3, 1^2),
 \end{array} \quad (5.52)$$

where the summation sign in (5.52) means direct sum.

For each $N \leq 7$, it was possible by this procedure to associate the irreducible representations uniquely with particular values of S . The distributions are given in appendix C. Therefore, essentially the same data is used to find μ_r'' as was required in (5.14) to determine μ_r . The susceptibility series for the various clusters is then obtained by making use of the identities in appendix A giving the λ_r'' in terms of the μ_s'' with $s \leq r$. Again the calculation was performed on the computer.

CHAPTER VI

DISCUSSION AND ANALYSIS OF SERIESA. Discussion of Series

For the spin one model of ferromagnetism defined by (3.2), using the procedures outlined in (4.28-4.35) together with the group theoretic simplifications described in the last chapter, it is possible to determine exactly the coefficients of K^r for $r \leq 7$ in the high temperature series expansions of the zero field partition function and the initial reduced susceptibility. The series $\phi_i(K)$ and $\psi_i(K)$, the weights associated with the cluster C_i in the partition function and susceptibility series respectively, are reproduced in appendix D together with all the linear combinations embodied in (4.30) and illustrated for a few cases in (4.32).


A number of features of the series is worth noting. While it is necessary to consider clusters containing a maximum of seven bonds we do not need to perform calculations on all such graphs. For certain classes of clusters containing b bonds, all coefficients of K^r in $\phi_i(K)$ or $\psi_i(K)$ are zero for $r < b + \lambda$ where the positive integer λ is known as the entry parameter. Thus, for example, all seven bonded clusters for which both $\phi_i(K)$ and $\psi_i(K)$ have $\lambda = 1$ need not be considered. Similarly all six-bonded clusters with $\lambda = 2$ in both cases make no contribution up to


K^7 and so on. The entry parameter of a cluster may not be the same for both the $\phi_i(K)$ and the $\psi_i(K)$ series and, as we see in the appendix D, for many clusters, only $\psi_i(K)$ was calculated.

To see how we can determine the entry parameter of any cluster, we recall how the various terms in the series $\phi_i(K)$ and $\psi_i(K)$ are associated with graphs of the same basic type as C_i . In the case of the $\phi_i(K)$, if C_i contains b bonds the coefficient of K^b or the primary weight of $\phi_i(K)$, as it is called, is given by the trace of spin operators corresponding to C_i itself. In our discussion of the direct method we noted how graphs with at least one vertex of degree one had zero weight i.e. the corresponding spin variable products were traceless. Hence we can conclude that all clusters containing one vertex of degree one will have no primary weight and an entry parameter of one in the corresponding $\phi_i(K)$. For example, as inspection of appendix D will show, $\phi_i(K)$ cor-

responding to the cluster  has no primary weight. In this

case the first non-zero coefficient, that of K^5 , will be associated with

the multiline graph . Similarly graphs with two vertices of


degree one e.g.  will have no primary weight or secondary

weight and the first non-zero coefficient, which occurs for K^7 , is associ-

ated with the multiline graph . Indeed for any cluster C_i

we can say that the entry parameter of its $\phi_i(K)$ is at least equal to the number of vertices of degree one in the cluster.

For the $\psi_i(K)$, however, the primary weight corresponds to the cluster C_i decorated with two crosses, these crosses indicating the addition of S^Z variables on the sites they occupy. If the cross is placed on a vertex of degree one, then the product of spin variables on that site will no longer be traceless. As an example while $\phi_i(K)$ correspond-

ing to the cluster  has an entry parameter of two, $\psi_i(K)$

has a non-zero primary weight associated with the decorated cluster



. A simple extension of the arguments presented leads to

the conclusion that the entry parameter of $\psi_i(K)$ is at least two less than the number of vertices of degree one in C_i .

Inspection of the series in appendix D will show that these entry parameter rules for both the $\phi_i(K)$ and the $\psi_i(K)$ are verified by calculation in all cases. The reason for the 'at least' in the statements of both rules is that we have no guarantee that the primary weight of a graph, which we expect from our considerations to be non-zero, may not in fact turn out to be zero because of the particular topology of the graph.

For example the $\psi_i(K)$ corresponding to the clusters




etc. which we expect to have zero entry



parameters actually have no primary weights. However, we have enumerated our clusters on the basis that the entry parameters are either equal to or two less than the number of vertices of degree one to ensure that all those contributing up to K^7 are included.

An important feature of these rules is that they provide a significant check on our calculations. If any $\phi_i(K)$ or $\psi_i(K)$, whose calculation involves many of the $\phi_i(K)$ and $\psi_i(K)$ previously evaluated, does not follow the rules then we are assured a mistake has occurred. It is a necessary though not sufficient condition for the correctness of our calculation that these rules be followed in all cases.

Of all the clusters we considered, only the linear chain of seven bonds i.e.  contains eight vertices. To calculate the corresponding $\psi_i(K)$ by the cluster expansion method would require consideration of matrices of dimension 3^8 and the considerable labour in evaluating the irreducible representations etc. of Π_8 just for the sake of one graph. To avoid all these difficulties we note only the primary

weight of the $\psi_i(K)$ given graphically by  has to

be evaluated and this can be done directly for all linear chains. For a linear chain of b bonds, the coefficient in the corresponding $\psi_i(K)$ of $K^b/3^b b!$ is given from (4.12) and (4.35) by

$$\frac{2}{3} \sum \text{trace} \left[\left(P_{12} - \frac{1}{3} \right) \left(P_{23} - \frac{1}{3} \right) \dots \left(P_{b,b+1} - \frac{1}{3} \right) S_1^z S_{b+1}^z \right] \quad (6.1)$$

where the summation in (6.1) is over all the $b!$ arrangements of the b brackets. Let us first consider the product $P_{12}P_{23}P_{34} \dots P_{b,b+1}$. In the direct product representation the only non-zero diagonal elements of this permutation operator correspond to the completely symmetric states given symbolically by $\alpha(1)\alpha(2) \dots \alpha(b+1)$, $\beta(1)\beta(2) \dots \beta(b+1)$ and $\gamma(1)\gamma(2) \dots \gamma(b+1)$ i.e. there are three diagonal elements equal to unity. The eigenvalues of $S_1^z S_{b+1}^z$ in these states are $+1$, 0 $+1$ respectively so that

$$\text{trace}(P_{12}P_{23}P_{34} \dots P_{b,b+1} S_1^z S_{b+1}^z) = 2 \quad (6.2)$$

All the $b!$ arrangements of permutation operators in (6.2) will have the same trace so that the contribution to (6.1) of this product is $4b!/3$. By similar arguments to those given above, we can show that all other contributions to the expansion of (6.1) in terms of products of permutation operators is zero. Each product contains a part of the form,

$$P_{12}P_{23} \dots P_{r,r+1} S_1^z, \quad (6.3)$$

where the number of permutation operators varies from zero to $b-1$. In the direct product representation, the permutation product in (6.3) has, as before, three non-zero diagonal elements each equal to unity. With the inclusion of the single S^z matrix, (6.3) then becomes traceless.

We therefore conclude that the coefficient of $K^b/3^{bb!}$ in $\psi_1(K)$

corresponding to the linear chain of b bonds is given by $4b!/3$. This result agrees with all our cluster expansion calculations for $b \leq 6$ and, as shown in appendix D, we conclude that for $b = 7$ the coefficient is equal to 6720.

The results described above have been combined according to (4.28) and (4.35) to obtain the logarithm of the partition function per site and the reduced initial susceptibility per site respectively on the face-centred cubic lattice. The reason for choosing this particular lattice is that, because of its high coordination number (twelve), so many configurations of spins occur for each power of K that the convergence of the series is expected to be most rapid for this lattice.

If we write each thermodynamic quantity in the form,

$$X = \sum_{n=0}^{\infty} a_n \frac{K^n}{3^n n!} = \sum_{n=0}^{\infty} b_n K^n, \quad (6.4)$$



the coefficients a_n for $\log Z/N$ and $kT\chi_0/Nm^2$ are reproduced in Table 6.1. Also included in that table are the series coefficients a_n for the specific heat per site C/Nk and the entropy per site in the form $(S_{\infty}-S)/Nk$ obtained from $\log Z/N$ by using (3.6) and (3.5) respectively. ($S_{\infty}/Nk = \log_e 3$ is the entropy per site in the limit of infinite temperature.)

Many of the lattice constants ($C_1; L$) required in the derivation of the above series are listed in Domb (1960). For the last term in


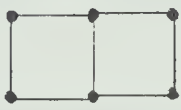
High temperature expansion coefficients for various thermodynamic quantities for the spin one exchange model on the face centred cubic lattice.

Thermodynamic quantity	a_0	a_1	a_2	a_3	a_4	a_5	a_6	a_7
$(\chi_{\text{MZ}})/N$	$\ell n 3$	0	48	288	-1440	-113,200	2,282,400	146,966,400
$kT\chi_0/Mn^2$	4/6	8	160	4656	182,160	8,865,408	511,455,744	34,231,956,480
$(S_{\infty}-S)/Nk$	0	0	48	576	-11320	-172,800	11,412,000	881,798,400
C/Nk	0	0	96	1728	-117,280	-864,000	68,472,000	6,172,588,800

the susceptibility series, however, thirty of the contributing clusters do not have their lattice constants listed in the literature. All of these were open configurations and their constants can be obtained by the 'symbolic equation' method described by Domb (1960). As an example

let us consider the cluster  and represent its lattice constant on the face-centred cubic lattice by (). It

can be formed from the hexagon by adding a single bond to any one of its vertices which can be done a total of sixty distinct ways. However, this added bond may be a chord or diagonal of the hexagon so that the

clusters  and  may be formed. the

number of times per site that these two configurations occur is twice the corresponding lattice constants since the added bond could have been originally attached to either of the two vertices it joins. We therefore set up the following symbolic equation,

$$\left(\text{hexagon with external bond} \right) = 60 \left(\text{hexagon} \right) - 2 \left(\text{hexagon with chord} \right) - 2 \left(\text{hexagon with diagonal} \right), \quad (6.5)$$

linking the unknown lattice constant on the left-hand side to a linear combination of lattice constants for the face-centred cubic lattice listed in Domb (1960). Hence we obtain in this case that

$$\left(\text{hexagon with external bond} \right) = 51,468. \quad (6.6)$$

The numerical value of the lattice constants of all thirty required open configurations are listed in appendix E together with the symbolic equations from which they were derived.

B. Analysis of Series

To locate the singularities in the thermodynamic functions such as magnetic susceptibility etc. to which the series in table 6.1 represent approximations for $T_c < T < \infty$, two well known extrapolation techniques are used. Both the ratio method introduced by Domb and Sykes (1957) and the Padé approximant method resurrected by Baker (1961) are attempts to extrapolate to the singular behavior of the physical functions near the critical temperature.

In the ratio method, if the dominant singularity of a function $f(x)$ occurs at some value x_c , it is assumed that near x_c $f(x)$ is of the form,

$$f(x) \approx A(1 - \frac{x}{x_c})^{-\gamma} , \quad (6.7)$$

where A is a constant. Then the coefficients b_n in the series expansion of $f(x)$ in powers of x should tend in the limit of large n to the binomial coefficients in the expansion of (6.7). The limiting form of the ratio of coefficients is given in that case by,

$$\frac{b_n}{b_{n-1}} \approx \frac{1}{x_c} (1 + \frac{\gamma-1}{n}) . \quad (6.8)$$

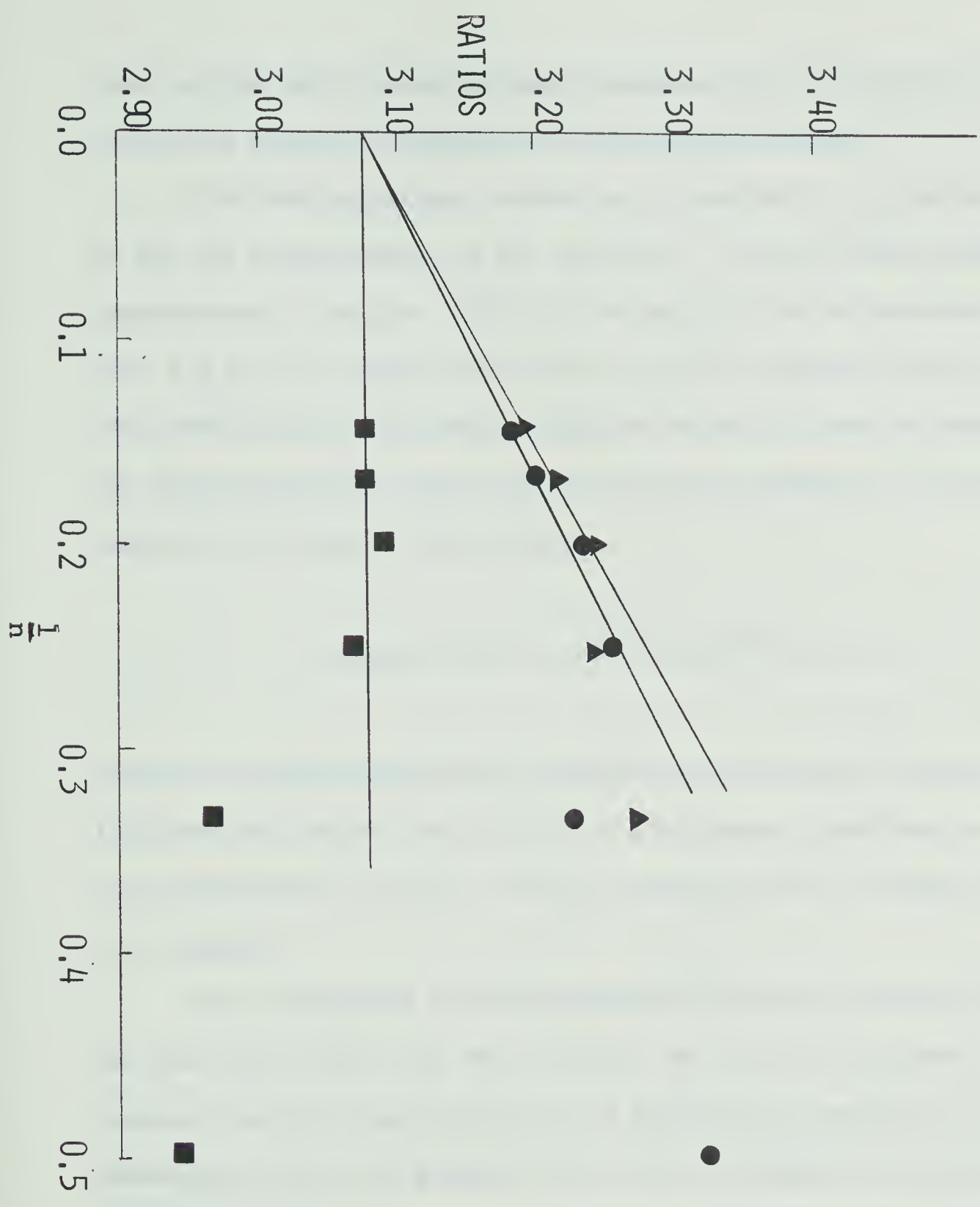
Therefore, if the ratio b_n/b_{n-1} is plotted against $1/n$, the graph should become linear for sufficiently large n . The projected intercept on the b_n/b_{n-1} axis will give $1/x_c$ and the limiting slope will equal $(\gamma-1)/x_c$. Similarly a plot of $(b_n/b_{n-2})^{1/2}$ against $1/n$ should approach linear behavior with intercept $1/x_c$ and slope $(\gamma-1)/x_c$. Also from (6.8) if we plot $nb_n/(n+\gamma-1)b_{n-1}$ against $1/n$, the graph should approach a horizontal line with an intercept of $1/x_c$. This last test is used if some value of $\gamma-1$ is postulated.

It is clear that this method can only be applied to a series of all positive coefficients. This means in our case that only the susceptibility series can be analysed by the ratio method. All three plots of the b_n given by (6.4) and Table 6.1 for the reduced susceptibility are shown in figure 6.1. For the first two, the linear region seems to be reached and we estimate that $1/K_c = 3.075 \pm 0.01$ or $K_c = 0.325 \pm 0.001$ and the critical index of the susceptibility $\gamma = 1.27 \pm 0.03$. There is a fashion in this field to postulate simple fractions for critical indices and if we yield to this and postulate $\gamma = 5/4$, the third plot of $nb_n/(n+\frac{1}{4})b_{n-1}$ against $1/n$ does seem to approach linear behavior with an intercept agreeing within graphical accuracy with the values quoted above.

It should be emphasized that in the first instance it is a significant assumption to postulate the form (6.7) for the susceptibility in the critical region for $K < K_c$ (i.e. above the critical temperature). However, the remarkable accuracy of the predictions of such a simple tech-

Fig. 6.1

Ratio plots for the spin one exchange model initial susceptibility coefficients ● indicate b_n/b_{n-1} , ▲ indicate $(b_n/b_{n-2})^{\frac{1}{2}}$ and
■ indicate $nb_n/(n+\frac{1}{4})b_{n-1}$.



nique as the ratio method strongly suggests that (6.7) may in fact represent the asymptotic behavior of the susceptibility.

The Padé approximant method can be applied to any series whether or not the coefficients are all positive. The $[N,M]$ Padé approximant approximates a function $f(x)$ by the ratio of two polynomials $P(x) = \sum_r p_r x^r$ of degree N and $Q(x) = \sum_s q_s x^s$ of degree M with $q_0 = 1$. The coefficients of the polynomials are chosen to make the expansion of the $[N,M]$ approximant agree exactly with the expansion of $f(x)$ to the first $M+N+1$ terms. The identity,

$$f(x)Q(x) - P(x) = ax^{M+N+1} + bx^{M+N+2} + \dots, \quad (6.9)$$

defines the approximant and by equating coefficients of powers of x in (6.9) one can solve a set of $M+N+1$ simultaneous equations to obtain the coefficients p_r and q_s . Such an operation can be readily performed on a computer.

The convergence of Padé approximants has been studied by Baker, Gammel and Wills (1961) and Baker (1965). We will not consider the problem here except to say that there is considerable evidence not all mathematically rigorous to suggest that in most problems of the type we are considering they converge very well when sufficient terms of the series are known (Fisher 1965).

The Padé approximant can be applied in the following four situations.

a) If the function $f(x)$ is of the form (6.7), then it is convenient to examine the approximants to the logarithmic derivative of the series for $f(x)$ given by,

$$\frac{d}{dx} \log f(x) \approx \frac{\gamma}{x_c - x} \quad , \quad (6.10)$$

which now has a simple pole. We expect the smallest positive zero in the denominator of the approximant to correspond to x_c and that the residue at that point will give the critical index γ .

b) If some precise independent estimate of the critical point x_c is known, then evaluating the approximants of the transformed series (6.10) multiplied by $(x_c - x)$ at $x = x_c$ should yield further estimates of γ .

c) If $f(x)$ is the form (6.7) and some independent estimate of γ is known, then

$$f(x)^{\frac{1}{\gamma}} \approx \frac{A^{\frac{1}{\gamma}}}{(1 - \frac{x}{x_c})} \quad (6.11)$$

and the smallest positive zero in the denominators of the approximants to (6.11) should correspond to x_c . Also the residues at that point should reproduce $A^{1/\gamma}$.

d) Baker et al. (1967) have suggested that the approximants to the expression ,

$$\frac{d}{dx} \log\left(\frac{df}{dx}\right) / \frac{d}{dx} \log f(x) = 1 + \frac{1}{\gamma} \quad , \quad (6.12)$$

evaluated at $x = x_c$ reproduce values of γ which are very insensitive to the choice of x_c .

All four procedures were applied to the susceptibility series. In case a) the results were inconclusive. For the $[M,N]$ approximants for which $M+N=6$, the values of K_c varied from 0.320 to 0.329 and γ from 1.17 to 1.39. While they are consistent with the much more accurate ratio method results, the scatter is much too large to draw any definite conclusions. Considerable improvement, however, occurs in method b) if we use the value of $K_c = 0.325$ suggested by the ratio method. The values of γ given by all the approximants for which $M+N = 5$ and 6 are reproduced in Table 6.3. Here the value of γ indicated is 1.27 ± 0.02 , provided we regard the value of the $[3,3]$ approximant as spurious. It is a matter of experience that occasionally one or two approximants yield results very different from all others. This can often occur if there exists in those approximants another singularity close to the physically relevant one. This value of γ , then, is impressively consistent with the independent estimate of the ratio method.

In method c), we chose the value of γ to be $5/4$, and the roots of the resulting approximants for $M + N = 5, 6$ and 7 are reproduced in Table 6.2. There is very little scatter in these results which yield a mean value of $K_c = 0.325 \pm 0.0005$. The mean value of the residues suggests that $A = 0.230 \pm .01$.

Because of the double differentiation involved in method d), it is more suitable for longer series, than we have available in this case.

Table 6.2

The $[M, N]$ Padé approximant estimates of K_c from $(\chi_0)^{4/5}$

M/N	1	2	3	4	5	6	7
0					0.3233	0.3252	0.3244
1				0.3233	0.3235	0.3246	
2			0.3259	0.3252	0.3245		
3		0.3219	0.3252	0.3273			
4	0.3221	0.3230	0.3243				
5	0.3249	0.3247					
6	0.3247						

Table 6.3

The $[M, N]$ Padé approximant estimates of γ from $[(K_c - K) \frac{d}{dK} \log \chi_0]_{K=K_c}$
with $K_c = 0.3252$

M/N	1	2	3	4	5	6
0					1.28	1.27
1				1.29	1.27	
2			1.24	1.27		
3		1.25	1.41			
4	1.28	1.27				
5	1.26					

It is not surprising that for $K_c = 0.325$, the values of γ obtained ranged from 1.17 to 1.39 for the approximants for which $M + N = 5$. It is interesting to note, however, that the mean value of the estimates gives $\gamma = 1.27$.

The very close agreement between the results of these two independent extrapolation techniques suggests strongly that just above the critical temperature the susceptibility is of the form (6.7) with the two main critical parameters given by,

$$\left. \begin{aligned} K_c &= 0.325 \pm 0.001 \\ \gamma &= 1.27 \pm 0.03 \end{aligned} \right\} . \quad (6.13)$$

We emphasize that the errors bars in (6.13) are not based on rigorous mathematics but rather are confidence limits we have set on the results on the basis of the consistency we have obtained. Perhaps one of the disadvantages of the Padé approximant is that it is not possible to give rigorous mathematical bound to its rate of convergence except in very specialized circumstances. While the series we have obtained contain a relatively small number of terms, they incorporate a considerable amount of information about the details of the lattice and how the correlations begin to increase as the temperature approaches critical. This is reflected in the contribution of larger graphs to the higher order terms. When regarded from this point of view, the accuracy of the results (6.13) is perhaps not so unreasonable and a good indication of how

the ultimate critical behavior is already incorporated in these truncated series.

As has been found for most other models, similar analysis of the specific heat series yields no consistent results. This is perhaps a reflection of the fact that the specific heat singularity, if it exists, is much weaker than for the susceptibility and requires much longer series to predict it precisely. Only very recently, for the three-dimensional Ising model have precise predictions of the specific heat behavior been made on the basis of a series with fourteen terms (Hunter 1967). Again Baker et al. (1967) on the basis of considerations we shall not discuss here have indirectly predicted the critical behavior of the specific heat for the spin one half Heisenberg model. They find that the specific heat is not even infinite at the critical temperature but rather has an infinite slope. If such behavior is characteristic of the specific heat in quantum mechanical models, it is not surprising that with only six terms we can not obtain any consistency.

It is possible however to evaluate the Pade approximants to the entropy series to find the fraction of the entropy change occurring above T_c . This is an experimentally interesting quantity which like the critical index is independent of the interaction constant J . The mean value we obtained is given by,

$$\frac{S_{\infty} - S_c}{S_{\infty}} = 0.33 \pm 0.02 . \quad (6.14)$$

The principal results of our calculation on the spin one exchange model are displayed in comparison with other relevant models in Table 6.4. Many of these results can be found in the review article of Domb and Miedema (1964). It is interesting to note that the spin one exchange model becomes disordered at a lower temperature than the spin one Heisenberg model, as is to be expected from the higher degeneracy of the ground state of a pair of spins interacting with the pure exchange interaction. The fraction of the total entropy change above T_c is directly comparable among the various models. We see that the Ising model differs markedly from all the quantum mechanical models in this respect, whereas the $(S_\infty - S_c)/S_\infty$ for the spin one-half and spin one exchange models are equal within 'experimental' error.

The quantity of greatest theoretical interest is probably the critical index γ which is expected to be independent of lattice structure. Here there is a clear difference between the spin one half and the spin one exchange models, with the value for the spin one Heisenberg model (the least well known) lying in between. However, the similarity in values of γ for the spin one exchange model and the spin one half Ising Model, two totally different models, suggests that we cannot always distinguish between quantum and classical models on the basis of susceptibility behavior alone.

As far as real spin one ferromagnets are concerned, we expect the inclusion of biquadratic terms in their interaction to lower their critical temperatures and reduce the value of the critical index γ in compari-

Table 6.4

Comparison of critical values for three lattice models.

Critical Quantity	Spin $\frac{1}{2}$ Ising Model	Spin $\frac{1}{2}$ Heisenberg or Exchange Model	Spin 1 Heisenberg Model	Spin 1 Exchange Model
kT_c/J	4.90	4.07	12.0	3.07
γ	5/4	1.43	1.38	1.27
$(S_\infty - S_c)/S_\infty$.147	.317	.27	.33

son with the pure spin one Heisenberg ferromagnets. It seems then that the details of the intermolecular interaction can produce detectable differences in critical behavior. However, the main question which still remains unanswered is precisely what features of the model are responsible for all the differences and similarities particularly when comparisons between quantum mechanical and classical models are made.

PART II

THE FREQUENCY-DEPENDENT INITIAL PERPENDICULAR
SUSCEPTIBILITY OF AND THE INELASTIC NEUTRON
SCATTERING FROM THE TWO DIMENSIONAL ISING MODEL

CHAPTER VII

THE PERPENDICULAR SUSCEPTIBILITY OF THE ISING MODELA. Introduction

The Ising model is one of the simplest and most studied models which exhibits a critical phase transition. In the presence of a magnetic field H , its Hamiltonian can be written in the form,

$$\mathcal{H} = -J \sum_{nn} \sigma_i \sigma_j - mH \sum_i \sigma_i, \quad (7.1)$$

where σ is a two-valued variable (+1 or -1) and m is the magnetic moment associated with it. We shall be primarily concerned with the interpretation of σ as a spin variable but as we mention in chapter one there are several other interpretations. Clearly (7.1) is a much simpler system than we considered in Part I if only for the fact that there is no question of non-commuting variables occurring in the system. Until recent years much of the work on the Ising model had been concerned with evaluating its equilibrium properties and a number of excellent reviews exist in the literature (e.g. Kadanoff et al. 1967; Fisher 1965 and 1967; Domb 1960).

A notable contribution to the study of the time dependent features of the model was made by Glauber (1963) who made use of the

'master equation' approach which discusses the first order differential equation governing the time evolution of the probability function associated with any given state of the system. His theory involves the important assumption that the transition probability that any spin will jump from one its states to the other depends only on the interaction of the given spin with its nearest neighbours. This probability is expressed in terms of two parameters which simultaneously describe the interaction of the Ising system with an external heat bath and the interaction between the individual spins. It is determined in part by the detailed balancing condition obeyed by the equilibrium state of the model.

Glauber considers the problem in detail for the case of a closed N-member chain and with further approximations and assumptions, the validity of which we shall not discuss here, he obtains for instance a parameterized expression for the complex frequency dependent magnetic susceptibility $\chi(\omega)$ defined by the relation,

$$\bar{M}(t) = \chi(\omega) H_0 e^{-i\omega t}, \quad (7.2)$$

where $\bar{M}(t)$ is the magnetization of the system at time t and $H_0 e^{-i\omega t}$ is a sinusoidally time dependent applied magnetic field.

Our aim in the following calculation is also to obtain an expression for the frequency dependent susceptibility of the Ising model but the details of our problem and the techniques we use are quite different. In the first place, we notice that the magnetic moment M given by,

$$\underline{M} = m \sum_k \sigma_k , \quad (7.3)$$

commutes with the Ising Hamiltonian (7.1) and so an isolated Ising system has no dynamic properties. In Glauber's case it is the interaction with the external heat bath which produces the time dependent features.

However, if we introduce the quantum mechanical Ising Model defined by the Hamiltonian,

$$\mathcal{H} = -J \sum_{n.n.} \sigma_i^z \sigma_j^z - m \underline{H} \cdot \underline{M} , \quad (7.4)$$

where the σ 's are now the usual Pauli spin matrices, we see that the magnetic moment operator given by,

$$\underline{M} = \sum_k (m_x \sigma_k^x , m_y \sigma_k^y , m_z \sigma_k^z) , \quad (7.5)$$

does not commute with (7.4) and so is not a constant of the motion. In particular, if we specialize to the case of a perpendicular field H_x only and perpendicular magnetic moment M_x , (7.4) and (7.5) become

$$\begin{aligned} \mathcal{H} &= \mathcal{H}_0 + \mathcal{H}_1 \\ &= -J \sum_{nn} \sigma_i^z \sigma_j^z - m H_x \sum_i \sigma_i^x , \end{aligned} \quad (7.6)$$

and
$$M_x = m \sum_k \sigma_k^x, \quad (7.7)$$

and we have the model whose static properties were first investigated by Fisher (1963). He found exactly for the honeycomb and plane square lattices the initial perpendicular susceptibility defined by,

$$\chi_{\perp}(T) = \lim_{H_x \rightarrow 0} \frac{\partial \bar{M}_x}{\partial H_x}, \quad (7.8)$$

where \bar{M}_x is the magnetization of the system in the x direction.

In our case, we are concerned with the dynamic or frequency dependent initial perpendicular susceptibility defined by (7.8) where H_x is now a sinusoidally time-dependent magnetic field of the form,

$$H_x(t) = H_x e^{-i\omega t}. \quad (7.9)$$

For an isolated Ising system, we shall apply the Kubo linear response theory (1957) to obtain an exact expression for the frequency dependent perpendicular susceptibility without the introduction of additional parameters. Detailed consideration is given to the honeycomb and plane square lattices and comparison with Fisher's static results leads to interesting conclusions regarding the susceptibility of an isolated system as opposed to that of a system in thermal contact with a heat bath.

B. The Kubo Formalism

The Kubo formalism is chosen in this problem because it is a particularly convenient way to study the response of a system to an external mechanical disturbance such as the perpendicular magnetic field in (7.6). As first pointed out by Kubo (1957) it can be applied to a variety of different physical problems but for our purposes it will be sufficient to develop the general formalism in the language of magnetism.

We make the initial assumption that our magnetic system in the remote past is in thermal equilibrium with its surroundings without the external magnetic field. If then a weak time dependent field $H(t)$ is introduced, the magnetization $\bar{M}(t)$ will be linear in the field and taking into account possible time lag effects we write

$$\bar{M}_{\mu}(t, T) = \sum_{\nu} \int_{-\infty}^t dt' \phi_{\mu\nu}(t-t', T) H_{\nu}(t') \quad , \quad (7.10)$$

where $\mu, \nu = x, y, z$ and $\phi_{\mu\nu}(t, T)$ is known as the response function. T is the temperature and t and t' the time. It is important to note that (7.10) is reasonable only for weak magnetic fields and Kubo's linear response theory based on it cannot be expected to yield accurate results with large fields. However, in our case we are interested in the zero field limit for which we expect (7.10) to be asymptotically exact. The response function characterizes then the temporal behavior of our system

and plays in this time dependent problem much the same role as the partition function does in static problems such as in Part I. As with the partition function, our main object is to calculate $\phi_{\mu\nu}(t, T)$ since it can be directly related to the experimentally significant macroscopic variables of the system.

In particular, to obtain an expression for the frequency dependent susceptibility, we consider the response of the system to a sinusoidal field of the form,

$$\underline{H}(t) = \underline{H}e^{-i\omega t + \delta t} \quad . \quad (7.11)$$

Substitution of (7.11) in (7.10) gives

$$\bar{M}_{\mu}(t, T) = \sum_{\nu} H_{\nu} \int_{-\infty}^t dt' e^{-i(\omega + i\delta)t'} \phi_{\mu\nu}(t - t', T) \quad (7.12)$$

$$= \sum_{\nu} H_{\nu} e^{-i(\omega + i\delta)t} \int_0^{\infty} ds \phi_{\mu\nu}(s, T) e^{i(\omega + i\delta)s} \quad (7.13)$$

$$= \sum_{\nu} \chi_{\mu\nu}(\omega, T) H_{\nu}(t) \quad , \quad (7.14)$$

where the frequency dependent susceptibility $\chi_{\mu\nu}(\omega, T)$ is given by,

$$\chi_{\mu\nu}(\omega, T) = \int_0^{\infty} dt \phi_{\mu\nu}(t, T) e^{i(\omega + i\delta)t} \quad . \quad (7.15)$$

The infinitesimal positive constant δ has been introduced to satisfy our initial condition of $H(t = -\infty) = 0$. We always finally take the limit $\delta \rightarrow +0$. (7.15) indicates that the frequency dependent susceptibility is simply the one-sided fourier transform of the response function.

To obtain $\phi_{\mu\nu}(t, T)$ we first identify the magnetization $\bar{M}(t, T)$ of our system at time t with the statistical average of \underline{M} , the magnetic moment operator, i.e.

$$\bar{M}(t, T) = \text{trace}[\rho(t, T)\underline{M}] \quad , \quad (7.16)$$

with \underline{M} given by (7.5). $\rho(t, T)$, the time dependent density matrix of the system, is obtained by solving the von Neumann equation of motion, namely,

$$\frac{d\rho(t, T)}{dt} = -\frac{i}{\hbar} [\mathcal{H}(t), \rho(t, T)] \quad , \quad (7.17)$$

where $\mathcal{H}(t)$ is the total Hamiltonian of the system and the following initial conditions are satisfied;

$$\left. \begin{aligned} H(t = -\infty) &= 0 \\ \rho(t = -\infty, T) &= \rho_{\text{eq}} = \frac{e^{-\beta\mathcal{H}_0}}{\text{trace}(e^{-\beta\mathcal{H}_0})} \end{aligned} \right\} . \quad (7.18)$$

\mathcal{H}_0 is the Hamiltonian of the system in the absence of the external field

$H(t)$ i.e. we write

$$\mathcal{H}(t) = \mathcal{H}_0 + \mathcal{H}_1(t) \quad (7.19)$$

with
$$\mathcal{H}_1(t) = - \underline{M} \cdot \underline{H}(t) \quad . \quad (7.20)$$

In that case, (7.17) becomes

$$\frac{d\rho(t,T)}{dt} = - \frac{i}{\hbar} [\mathcal{H}_0, \rho(t,T)] - \frac{i}{\hbar} [\mathcal{H}_1(t), \rho(t,T)] , \quad (7.21)$$

and since we are interested only in the linear response of the magnetization to the field, we replace $\rho(t,T)$ in the second term on the right hand side of (7.21) by ρ_{eq} given in (7.18) so that

$$\frac{d\rho(t,T)}{dt} \approx - \frac{i}{\hbar} [\mathcal{H}_0, \rho(t,T)] - \frac{i}{\hbar} [\mathcal{H}_1(t), \rho_{eq}] \quad . \quad (7.22)$$

Hence we obtain the solution,

$$\rho(t,T) = \rho_{eq} - \frac{i}{\hbar} \int_{-\infty}^t dt' e^{-i\mathcal{H}_0(t-t')/\hbar} [\mathcal{H}_1(t'), \rho_{eq}] e^{i\mathcal{H}_0(t-t')/\hbar} , \quad (7.23)$$

which can be verified directly by substitution in (7.22).

Finally to obtain an expression for the response function, we substitute (7.23) in (7.16) and comparing the result with (7.10) we can

make the identification,

$$\phi_{\mu\nu}(t-t', T) = \frac{i}{\hbar} \text{trace} \left\{ e^{-iH_0(t-t')/\hbar} [M_\nu, \rho_{eq}] e^{iH_0(t-t')/\hbar} M_\mu \right\}, \quad (7.24)$$

provided

$$\text{trace}(\rho_{eq} M_\mu) = 0. \quad (7.25)$$

In our case where $\mu = x$ and H_0 is given by (7.6), (7.25) is certainly satisfied.

(7.24) is the important relation we require to link the macroscopic variables of our system to the microscopic dynamics as provided by the Hamiltonian. Using the invariance property of the trace under cyclic transformation, namely,

$$\text{trace}(ABC \dots Z) = \text{trace}(ZAB \dots Y) \quad \text{etc.}, \quad (7.26)$$

and setting $t' = 0$, (7.24) can be more compactly written in the form,

$$\phi_{\mu\nu}(t, T) = \frac{i}{\hbar} \text{trace} \left\{ [M_\nu, \rho_{eq}] M_\mu(t) \right\} \quad (7.27)$$

$$= \frac{i}{\hbar} \text{trace} \left\{ M_\nu \rho_{eq} M_\mu(t) - \rho_{eq} M_\nu M_\mu(t) \right\} \quad (7.28)$$

$$= \frac{i}{\hbar} \text{trace} \left\{ \rho_{eq} (M_\mu(t) M_\nu - M_\nu M_\mu(t)) \right\} \quad (7.29)$$

$$= \frac{i}{\hbar} \left\langle [M_\mu(t), M_\nu] \right\rangle . \quad (7.30)$$

In (7.27 - 7.30) we have

$$M_\mu(t) = e^{iH_0 t/\hbar} M_\mu e^{-iH_0 t/\hbar} . \quad (7.31)$$

In conclusion, we should note from (7.30) one of the important features of Kubo's linear response theory. It expresses a time-dependent property of the system, namely the response function, as a statistical average of operators for the equilibrium system i.e. in the absence of the external disturbance.

C. Application of the Formalism

To obtain the frequency dependent initial perpendicular susceptibility of the Ising model with the Hamiltonian given by (7.6), we first must evaluate (7.30) for the particular case of $\mu = \nu = x$ and then substitute in (7.15).

Firstly we note that

$$e^{iH_0 t/\hbar} = e^{i \frac{\omega_0 t}{2} \sum_{nn} \sigma_n^z \sigma_n^z} \quad (7.32)$$

$$(\omega_0 = 2J/\hbar)$$

$$= \prod_{nn} e^{\mp i \frac{\omega_0 t}{2} \sigma_i^z \sigma_j^z} \quad (7.33)$$

$$= \prod_{nn} \left(\cos \frac{\omega_0 t}{2} \mp i \sigma_i^z \sigma_j^z \sin \frac{\omega_0 t}{2} \right), \quad (7.34)$$

where we have used the identity,

$$(\sigma^z)^{2n} = I, \quad (7.35)$$

for n any positive integer. (7.34) is completely analogous to the familiar van der Waerden identity,

$$e^{-\beta \mathcal{H}_0} = \prod_{nn} (\cosh K + \sigma_i^z \sigma_j^z \sinh K), \quad (7.36)$$

$$(K = J/kT)$$

used in the expansion of the Ising model partition function.

Substituting (7.34) in (7.30) for $\mu = \nu = x$ and using (7.7), we obtain

$$\phi_{xx}(t, T) = \frac{im^2}{\hbar} \left(\cos \frac{\omega_0}{2} t \right)^{Nq} \sum_{i', j'} \left\langle \prod_{nn} \left(1 - i \sigma_i^z \sigma_j^z \tan \frac{\omega_0}{2} t \right) \right\rangle$$

$$\sigma_1^x, \prod_{nn} (1 + i\sigma_k^z \sigma_l^z \tan \frac{\omega_0 t}{2}), \sigma_{j'}^x] \rangle \quad (7.37)$$

where q is the coordination number of the lattice and N the number of lattice sites. Using the familiar anticommutation properties of the Pauli spin operators, (7.37) becomes

$$\phi_{xx}(t, T) = \frac{\hbar^2}{\hbar} (\cos \frac{\omega_0}{2} t)^{Nq} \sum_{i', j'} \left\langle \left[\prod_{nn} (1 - i\sigma_i^z \sigma_j^z \tan \frac{\omega_0}{2} t) \times \right. \right. \quad (7.38)$$

$$\left. (1 + i\sigma_i^z \sigma_j^z \tan \frac{\omega_0}{2} t) \prod_{k=1}^q (1 - i\sigma_k^z \sigma_{i'}^z \tan \frac{\omega_0}{2} t) \sigma_{i'}^x, \sigma_{j'}^x \right] \rangle$$

$$= \frac{\hbar^2}{\hbar} (\cos \frac{\omega_0}{2} t)^{Nq} \sum_{i', j'} \left\langle \left[(1 + \tan^2 \frac{\omega_0}{2} t)^{(\frac{Nq}{2} - q)} \times \right. \right.$$

$$\left. \prod_{k=1}^q (1 - \tan^2 \frac{\omega_0}{2} t - 2i\sigma_k^z \sigma_{i'}^z \tan \frac{\omega_0}{2} t) \sigma_{i'}^x, \sigma_{j'}^x \right] \rangle. \quad (7.39)$$

Making use of the following simple trigonometric identities,

$$a) \quad 1 + \tan^2 \frac{\omega_0}{2} t = \sec^2 \frac{\omega_0}{2} t, \quad (7.40)$$

$$b) \quad u = \tan \omega_0 t = \frac{2 \tan \frac{\omega_0}{2} t}{1 - \tan^2 \frac{\omega_0}{2} t} , \quad (7.41)$$

$$c) \quad 1 - \tan^2 \frac{\omega_0}{2} t = \cos \omega_0 t \sec^2 \frac{\omega_0}{2} t , \quad (7.42)$$

we obtain

$$\phi_{xx}(t, T) = \frac{\hbar^2}{\hbar} (\cos \omega_0 t)^q \sum_{i', j'} \left\langle \left[\prod_{k=1}^q (1 - i \sigma_k^z \sigma_{i'}^z, u) \sigma_{i'}^x, \sigma_{j'}^x \right] \right\rangle \quad (7.43)$$

$$= \frac{\hbar^2}{\hbar} (\cos \omega_0 t)^q \sum_{i', j'} \left\langle \left[\prod_{k=1}^q (1 - i \sigma_k^z \sigma_{i'}^z, u), \sigma_{j'}^x \right] \sigma_{i'}^x \right\rangle \quad (7.44)$$

where in obtaining (7.44) we have made use of the commutator identity,

$$[AB, C] = A[B, C] + [A, C]B . \quad (7.45)$$

The commutator in (7.44) can obviously be non-zero in only two cases; a) $i' = j' = j$ and b) i' and j' are nearest neighbour sites. Considering the latter case, we get

$$\left[\prod_{k=1}^q (1 - i \sigma_k^z \sigma_{i'}^z, u), \sigma_{j'}^x \right] \sigma_{i'}^x$$

$$= \prod_{k \neq j'} (1 - i\sigma_k^z \sigma_{i'}^z u) [(1 - i\sigma_{j'}^z \sigma_{i'}^z u), \sigma_{j'}^x] \sigma_{i'}^x \quad (7.46)$$

$$= - \left\{ \prod_{k \neq j'} (1 - i\sigma_k^z \sigma_{i'}^z u) \right\} 2i\sigma_{j'}^y \sigma_{i'}^z \sigma_{i'}^x u \quad (7.47)$$

We notice in (7.47) that $\sigma_{j'}^y$ is the only variable at the j' 'th site.

Thus when the thermal average is taken to obtain the response function i.e. when ρ_{eq} is included, only terms of the form $(\sigma_{j'}^z)^n \sigma_{j'}^y$ (n a positive integer) will occur at the j' 'th site. For the Pauli spin operators, all such terms are traceless. The thermal averaging involves taking the trace over the direct product space of all N spins which will be zero if the trace over any one spin is zero. Case b) then gives no contribution and (7.44) can now be written as,

$$\phi_{xx}(t, T) = \frac{im^2}{\hbar} (\cos \omega_0 t)^q \sum_j \left\langle \left[\prod_{k=1}^q (1 - i\sigma_k^z \sigma_j^z u), \sigma_j^x \right] \sigma_j^x \right\rangle. \quad (7.48)$$

Using a generalisation of (7.45) we then obtain

$$\phi_{xx}(t, T) = \frac{im^2}{\hbar} (\cos \omega_0 t)^q \sum_j \left\langle \left\{ \sum_{k=1}^q \prod_{i=1}^{k-1} (1 - i\sigma_i^z \sigma_j^z u) \times \right. \right. \\ \left. \left. [(1 - i\sigma_k^z \sigma_j^z u), \sigma_j^x] \prod_{i=k+1}^q (1 - i\sigma_i^z \sigma_j^z u) \right\} \sigma_j^x \right\rangle \quad (7.49)$$

$$= \frac{2im^2}{\hbar} (\cos \omega_0 t)^{q-1} \sin \omega_0 t \sum_j \left\langle \left\{ \sum_{k=1}^q \prod_{i=1}^{k-1} (1 - i\sigma_i^z \sigma_j^z u) \sigma_k^z \sigma_j^y \times \right. \right. \\ \left. \left. \prod_{i=k+1}^q (1 - i\sigma_i^z \sigma_j^z u) \right\} \sigma_j^x \right\rangle. \quad (7.50)$$

We now expand the expression within the $\{ \}$ brackets in (7.50) in powers of u . Associated with u^r is a sum of distinct products of $r+1$ pairs of spin operators of which r are z, z pairings and one is of the form $\sigma_k^z \sigma_j^y$. To enumerate all the products we first of all consider the ${}^q C_{r+1}$ distinct selections of $r+1$ nearest neighbour sites to the j th site. Corresponding to each selection there will be $r+1$ distinct products of the form

$$(\sigma_{k_1}^z \sigma_j^z)(\sigma_{k_2}^z \sigma_j^z) \dots (\sigma_k^z \sigma_j^y) \dots (\sigma_{k_{r+1}}^z \sigma_j^z), \quad (7.51)$$

each differing only in the location of the $\sigma_k^z \sigma_j^y$ pair. Here $k_1 k_2 \dots k_{r+1}$ represents a particular selection of $r+1$ nearest neighbour sites.

(7.51) can be rearranged to become,

$$\sigma_{k_1}^z \sigma_{k_2}^z \dots \sigma_k^z \dots \sigma_{k_{r+1}}^z (\sigma_j^z)^\alpha \sigma_j^y (\sigma_j^z)^\beta, \quad (7.52)$$

where

$$\alpha + \beta + 1 = r + 1. \quad (7.53)$$

If we examine the product of spin variables on the j th site in (7.52) and recall the anticommutation properties of the σ 's, we see that the $r+1$ distinct products will cancel in pairs. In particular if $r+1$ is an even integer there will be no resultant contribution to the coefficient of u^r . For instance for the simple quadratic lattice for which $q=4$ if we take $r=1$, then the two products associated with one of the six possible choices of two sites from four would be given by $\sigma_1^z \sigma_2^z \sigma_j^z \sigma_j^y$ and $\sigma_1^z \sigma_2^z \sigma_j^y \sigma_j^z$ where we have arbitrarily labelled the two nearest neighbour vertices 1 and 2. These two terms anticommute and their sum is zero. Similarly the other five pairs of terms cancel so that the coefficient of u for the simple quadratic lattice is zero.

If, however, $r+1$ is an odd integer then there will always be one spin product uncanceled which we can choose to be of the form,

$$\sigma_{k_1}^z \sigma_{k_2}^z \dots \sigma_{k_{r+1}}^z (\sigma_j^z)^r \sigma_j^y = \sigma_{k_1}^z \sigma_{k_2}^z \dots \sigma_{k_{r+1}}^z \sigma_j^y, \quad (7.54)$$

using (7.35). For example for the simple quadratic lattice, there are four spin products contributing to the coefficient of u^2 in (7.50), namely,

$$\sigma_1^z \sigma_2^z \sigma_3^z \sigma_j^y, \sigma_1^z \sigma_2^z \sigma_4^z \sigma_j^y, \sigma_1^z \sigma_3^z \sigma_4^z \sigma_j^y, \sigma_2^z \sigma_3^z \sigma_4^z \sigma_j^y, \quad (7.55)$$

one each for the four possible selections of three nearest neighbour sites to the j th (labelled 1,2,3,4).

From all these considerations we can now write (7.50) in the form,

$$\phi_{xx}(t, T) = \frac{2iNm^2}{\hbar} (\cos \omega_0 t)^{q-1} \sin \omega_0 t \left(\sum_{\substack{r=0 \\ 2r+1 \leq q}} (-1)^r u^{2r} \times \right. \quad (7.56)$$

$$\left. \sum_k^C \langle \sigma_{k_1}^z \sigma_{k_2}^z \dots \sigma_{k_{2r+1}}^z \sigma_0^y \sigma_0^x \rangle \right)$$

$$= \frac{2Nm^2}{\hbar} (\cos \omega_0 t)^{q-1} \sin \omega_0 t \left(\sum_{\substack{r=0 \\ 2r+1 \leq q}} (-1)^r u^{2r} \times \right. \quad (7.57)$$

$$\left. \sum_k^C \langle \sigma_{k_1}^z \sigma_{k_2}^z \dots \sigma_{k_{2r+1}}^z \sigma_0^z \rangle \right),$$

where in (7.56) and (7.57) we have invoked the translational symmetry of the lattice to replace the summation over j by a factor of N and re-labelled the j th site as the zeroth. The first summation in (7.56) and (7.57) is over all positive integer values of r such that $2r+1 \leq q$. The second summation is over all possible selections of $2r+1$ sites from the q nearest neighbours to the zeroth.

The equation (7.57) is the desired exact relation between the response function and the equilibrium spin correlation functions, valid for all regular lattices. The form of the relationship depends only on

the coordination number, not on the particular lattice nor even on the dimensionality of the lattice. For two dimensional lattices all the required correlation functions are either known or can be evaluated by known methods (Montroll, Potts and Ward 1963). In the next chapter, the particular form of the response function will be determined for the honeycomb and square lattices and used in conjunction with (7.15) to obtain the frequency dependent initial perpendicular susceptibility of these two lattices.

CHAPTER VIII

APPLICATION TO HONEYCOMB AND PLANE SQUARE LATTICESA. Honeycomb Lattice

Putting $q = 3$ in (7.57) the response function for the honeycomb lattice is given by,

$$\phi_{xx}^H(t, T) = \frac{2Nm^2}{\hbar} \cos^2 \omega_0 t \sin \omega_0 t (3 \langle \sigma_0^z \sigma_1^z \rangle - \langle \sigma_0^z \sigma_1^z \sigma_2^z \sigma_3^z \rangle \tan^2 \omega_0 t) , \quad (8.1)$$

where spins 1, 2, and 3 are the nearest neighbours to spin 0. Taking the one-sided fourier transform of $\phi_{xx}^H(t, T)$ according to (7.15), we obtain the frequency dependent initial perpendicular susceptibility $\chi_{\perp}^H(\omega, T)$ in the form,

$$\begin{aligned} \frac{J\chi_{\perp}^H(\omega, T)}{Nm^2} &= \frac{1}{3} \left[\frac{3}{4} \langle \sigma_0 \sigma_1 \rangle + \frac{1}{4} \langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle \right] \frac{1}{1 - (\omega/3\omega_0)^2} \\ &+ \frac{3}{4} \left[\langle \sigma_0 \sigma_1 \rangle - \langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle \right] \frac{1}{1 - (\omega/\omega_0)^2} , \end{aligned} \quad (8.2)$$

where $\sigma = \sigma^z$. For the honeycomb lattice, (8.1) and (8.2) provide exact solutions when the known values of the correlations are inserted.

Any correlation of a finite number of spins in a two dimensional

Ising model can be obtained by the dimer-Pfaffian method (Mantroll, Potts and Ward 1963) which we will not discuss here. However, the nearest neighbour correlation $\langle \sigma_0 \sigma_1 \rangle$ is of course directly proportional to the energy and was first calculated for the honeycomb lattice by Houtappel (1950) to give

$$\langle \sigma_0 \sigma_1 \rangle = \frac{2}{3} [\coth 2K + \alpha \mathcal{K}(k_1)] , \quad (8.3)$$

in which $\mathcal{K}(k_1)$ is the complete elliptic integral of the first kind defined, for example, by Jahnke and Emde (1945) as,

$$\mathcal{K}(k_1) = \int_0^{\pi/2} \frac{dx}{\sqrt{1 - k_1^2 \sin^2 x}} . \quad (8.4)$$

Also

$$k_1^2 = \begin{cases} \frac{\cosh^2 K + 3 \sinh^2 K}{16 \sinh^6 K \cosh^2 K} & K > K_c \\ \frac{16 \sin^6 K \cosh^2 K}{\cosh^2 K + 3 \sinh^2 K} & K < K_c \end{cases} \quad (8.5)$$

and

$$\alpha = \begin{cases} \frac{\cosh 2K(\cosh 2K - 2)}{4\pi \sinh^4 K} & K > K_c \\ \frac{\sinh 4K(\cosh 2K - 2)(\cosh K)^{1/2}}{4\pi \sinh^2 K(\cosh 3K)^{1/2}} & K < K_c \end{cases} \quad (8.6)$$

where, again, $K = J/kT$.

The four particle correlation $\langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle$ is most conveniently calculated by using a transformation theorem developed by Fisher (1959). This theorem shows that a correlation function involving a spin σ_0 can be expressed as a linear combination of correlation functions involving the q spins $\sigma_1, \sigma_2, \sigma_3 \dots \sigma_q$, which are nearest neighbours to σ_0 . In our proof, we shall assume initially that the interaction constant J may not necessarily be the same for all bonds in the lattice. In that case, the correlation product between the spins $\sigma_0, \sigma_g, \sigma_h, \dots, \sigma_m$, is given by,

$$\langle \sigma_0 \sigma_g \sigma_k \dots \sigma_m \rangle = \frac{\text{trace} [\sigma_0 \sigma_g \sigma_k \dots \sigma_m e^{\sum_{ij} K_{ij} \sigma_i \sigma_j}]}{\text{trace } e^{\sum_{ij} K_{ij} \sigma_i \sigma_j}}, \quad (8.7)$$

where the trace is over the direct product space of all the spins in the lattice. Let us consider in particular the trace over the zeroth spin space in the numerator of (8.7) i.e.

$$T_0(\sigma_1, \sigma_2, \dots, \sigma_q) = \text{trace}_0 \left[\sigma_0 e^{(K_1 \sigma_0 \sigma_1 + K_2 \sigma_0 \sigma_2 + \dots + K_q \sigma_0 \sigma_q)} \right] \quad (8.8)$$

where we now regard $\sigma_1, \sigma_2, \sigma_3, \dots, \sigma_q$ as scalar parameters which can equal to +1 or -1. We may try to represent (8.8) in the form,

$$T_0(\sigma_1, \sigma_2, \dots, \sigma_q) = \left\{ \sum_{r=1}^q \alpha_r \sigma_r + \sum_{t=1}^q \sum_{a_t, b_t, c_t}^3 \beta_t \sigma_{a_t} \sigma_{b_t} \sigma_{c_t} \right. \quad (8.9)$$

$$\left. + \sum_{u=1}^q \sum_{a_u, b_u, c_u, d_u, e_u}^5 \gamma_u \sigma_{a_u} \sigma_{b_u} \sigma_{c_u} \sigma_{d_u} \sigma_{e_u} \right\} \text{trace}_0 e^{(K_1 \sigma_0 \sigma_1 + K_2 \sigma_0 \sigma_2 + \dots + K_q \sigma_0 \sigma_q)} ,$$

where the q coefficients α_r , the ${}_q C_3$ coefficients β_t , the ${}_q C_5$ coefficients γ_u , etc., are functions only of the q interaction parameters K_1, K_2, \dots, K_q between the spin σ_0 and its nearest neighbours $\sigma_1, \sigma_2, \dots, \sigma_q$ respectively. The summations in (8.9) are successively over all distinct selections of 1, 3, 5, etc. spins from the q nearest neighbours to σ_0 . Because (8.8) changes sign when the signs of the spins $\sigma_1, \sigma_2, \dots, \sigma_q$ are simultaneously altered, we do not need to consider combinations of even numbers of spins in (8.9). The number of unknown parameters in (8.9) is

$${}_q C_1 + {}_q C_3 + {}_q C_5 + \dots = 2^{q-1} . \quad (8.10)$$

There are 2^q distinct joint spin states of the nearest neighbour spins and the identity of (8.8) and (8.9) must hold for all of them. However, since both expressions change sign on simultaneous reversal of the q nearest neighbour spins, we obtain only $(1/2)2^q = 2^{q-1}$ independent equations between (8.8) and (8.9) which therefore will exactly determine the set of coefficients $\alpha_r, \beta_t, \gamma_u \dots$ ensuring that (8.9) is a faithful representation of (8.8). On inserting this transformation into (8.7), we get

$$\langle \sigma_0 \sigma_g \sigma_h \dots \sigma_m \rangle = \sum_{r=1}^q \alpha_r \langle \sigma_r \sigma_g \sigma_h \dots \sigma_m \rangle + \sum_{t=1}^q \sum_{c=3}^3 \beta_t \langle \sigma_{a_t} \sigma_{b_t} \sigma_{c_t} \sigma_g \sigma_h \dots \sigma_m \rangle + \dots, \quad (8.11)$$

which expresses the correlation involving σ_0 as a sum of correlation functions including the nearest neighbour spins $\sigma_1, \sigma_2, \dots \sigma_q$ in place of σ_0 .

We shall use this theorem to determine $\langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle$ in terms of the second neighbour correlation $\langle \sigma_1 \sigma_2 \rangle$ for the honeycomb lattice with a unique interaction constant. In that case (8.8) becomes

$$T_0(\sigma_1, \sigma_2, \sigma_3) = \text{trace}_0 [\sigma_0 e^{K(\sigma_0 \sigma_1 + \sigma_0 \sigma_2 + \sigma_0 \sigma_3)}] , \quad (8.12)$$

and from the symmetry of the lattice, (8.9) is

(8.13)

$$T_0(\sigma_1, \sigma_2, \sigma_3) = \left\{ \alpha(\sigma_1 + \sigma_2 + \sigma_3) + \beta(\sigma_1 \sigma_2 \sigma_3) \right\} \text{trace}_0 [e^{K(\sigma_0 \sigma_1 + \sigma_0 \sigma_2 + \sigma_0 \sigma_3)}] .$$

Now there are only two distinct joint states of the three nearest neighbour spins corresponding to a) $\sigma_1 = \sigma_2 = \sigma_3 = +1$ and b) typically $\sigma_1 = -1, \sigma_2 = \sigma_3 = +1$ which will be, however, sufficient to determine the two unknown parameters in (8.13). The identity of (8.12) and (8.13) for case a) leads to the equation,

$$3\alpha + \beta = \tanh 3K \quad , \quad (8.14)$$

and for case b) to

$$\alpha - \beta = \tanh K \quad . \quad (8.15)$$

Solving (8.14) and (8.15), substituting (8.13) in (8.7) and making use of (7.35) together with the symmetry of the lattice, we finally obtain

$$(8.16)$$

$$\langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle = \frac{3}{4} (\tanh 3K + \tanh K) \langle \sigma_1 \sigma_2 \rangle + \frac{1}{4} (\tanh 3K - 3 \tanh K) \quad .$$

Using an exactly similar procedure for $\langle \sigma_0 \sigma_1 \rangle$, we also get

$$\langle \sigma_1 \sigma_2 \rangle = \frac{1}{2} (\coth K + 3 \tanh K) \langle \sigma_0 \sigma_1 \rangle - \frac{1}{2} (1 + \tanh^2 K) \quad . \quad (8.17)$$

Substitution of (8.17) in (8.16) gives after some trigonometric simplification,

$$\langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle = \frac{3}{2} (1 + \tanh^2 K) \langle \sigma_0 \sigma_1 \rangle - \frac{1}{2} \tanh K (\tanh^2 K + 3) \quad . \quad (8.18)$$

(8.18) represents the exact known value of the four particle correlation since we have already stated the exact solution of $\langle \sigma_0 \sigma_1 \rangle$ in equations (8.3) to (8.6).

The most obvious check on $\chi_{\perp}^H(\omega, T)$ given in (8.2) is to compare the zero frequency limit with Fisher's (1963) static susceptibility result for the honeycomb lattice. Substituting (8.18) in (8.2) we find for $\omega = 0$ that

$$\frac{J\chi_{\perp}^H(0, T)}{Nm^2} = \frac{1}{3} \tanh K (\tanh^2 K + 3) - \tanh^2 K \langle \sigma_0 \sigma_1 \rangle. \quad (8.19)$$

Specializing Fisher's equation (4.17) to the honeycomb lattice and making use of (8.17), we find perfect agreement between our results and Fisher's.

The most striking feature of $\chi_{\perp}^H(\omega, T)$ consists of the two resonances occurring at $\omega = 2J/\hbar$ and $6J/\hbar$. It is easy to see the physical origin of these resonances. In the honeycomb lattice, a given spin will be found in one of four environments. In two environments, all three of its nearest neighbours are orientated either parallel or antiparallel to the central spin. In these configurations, the energy required to flip the central spin will be respectively $+6J$ and $-6J$ and the $\omega = 6J/\hbar$ resonance corresponds to an absorption of energy from the magnetic field by those spins in the all parallel state. In the other two environments, the central spin has either two of its neighbours parallel and one anti-

parallel or vice versa. Clearly the $\omega = 2J/k$ resonance occurs because of spins in the two parallel, one antiparallel configuration. It is interesting to note that for critical temperatures of the order of a few degrees Kelvin these resonances are in the microwave region.

It should be pointed out that the $\chi_{\perp}^H(\omega, T)$ which we have been calling the perpendicular susceptibility is in fact the real part of a complex frequency dependant susceptibility of which the imaginary part is given by,

$$\frac{J\chi_{\perp}^H(\omega, T)^I}{Nm^2} = \omega_0 \left[\left(\frac{3}{8} \langle \sigma_0 \sigma_1 \rangle + \frac{1}{8} \langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle \right) (\delta(3\omega_0 - \omega) - \delta(3\omega_0 + \omega)) \right. \\ \left. + \frac{3}{8} (\langle \sigma_0 \sigma_1 \rangle - \langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle) (\delta(\omega_0 - \omega) - \delta(\omega_0 + \omega)) \right], \quad (8.20)$$

and corresponds to the energy absorption by the spin system. The resonances in (8.20) are of zero linewidth because the only interactions in this model are the nearest neighbour Ising interactions. Introducing additional near neighbour interactions would simply increase the number of zero linewidth resonances. In a real crystal, of course, additional long range forces such as magnetic dipole-dipole interactions and spin-lattice forces will result in a finite linewidth to the resonances. However, if a crystal approximates an Ising lattice at least the location of the resonances should be predicted by this analysis.

Returning to consideration of (8.2), the real part of the fre-

quency-dependent susceptibility, it is interesting to examine the temperature dependence of the amplitude of the resonances. At $T = 0$, where all even correlations are unity, corresponding to the completely magnetized state, the resonance at $\omega = \omega_0$ in (8.2) disappears. This is because with all spins aligned the energy necessary to overturn a single spin can only be $+6J$. At the other extreme of $T \rightarrow \infty$, $\langle \sigma_0 \sigma_1 \rangle / \langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle \gg 1$, and the amplitudes of the $\omega = \omega_0$ and $\omega = 3\omega_0$ resonances are predicted by (8.2) to be in the ratio of 3 to 1. This simply corresponds to the relative probabilities in a random array of spins for the arrangement of two first neighbour spins parallel and one antiparallel to the arrangement of all spins parallel.

We shall also consider the critical region given by $\sinh^2 K_c^H = 1/2$ for the honeycomb lattice (Houtappel 1950). From (8.5) $K \sim K_c$ corresponds to $k_1^2 \sim 1$ and $\alpha \sim 0$ and, from Jahnke and Emde (1945), to

$$\mathcal{K}(k_1) \approx \log \frac{4}{\sqrt{1-k_1^2}} . \quad (8.21)$$

Expanding these terms in a Taylor series about $K = K_c$ and retaining only lowest order terms, we find

$$\langle \sigma_0 \sigma_1 \rangle \approx \frac{4\sqrt{3}}{9} - \frac{4\sqrt{3}}{3\pi} K_c^H \left(1 - \frac{T}{T_c^H}\right) \log \left| 1 - \frac{T}{T_c^H} \right| , \quad (8.22)$$

and substituting (8.22) and (8.18) in (8.2) that

$$\begin{aligned} & \frac{J}{Nm^2} [\chi_{\perp}^H(\omega, T) - \chi_{\perp}^H(\omega, T_c^H)] \\ & \approx \frac{\sqrt{3}}{\pi} K_c^H \left[\frac{1}{1-(\omega/\omega_0)^2} - \frac{5/9}{1-(\omega/3\omega_0)^2} \right] \left(1 - \frac{T}{T_c^H}\right) \log \left| 1 - \frac{T}{T_c^H} \right|. \end{aligned} \quad (8.23)$$

We see that for all frequencies $\chi_{\perp}^H(\omega, T)$ is finite in magnitude but has an infinite slope at T_c . In particular at low frequencies the slope is positive infinite. This behavior contrasts sharply with the much stronger singularity exhibited at T_c by the susceptibility in the spin one exchange model. This is very much related to the fact that the latter susceptibility can be shown to be equivalent to an infinite sum of spin correlation functions whereas from (8.2) and (8.18) the perpendicular susceptibility of the Ising model is related to only one correlation.

To illustrate the frequency and temperature dependence of the perpendicular susceptibility of the honeycomb lattice, the behavior of $\chi_{\perp}^H(\omega, T)$ as a function of temperature for various fixed values of ω approaching $\omega = \omega_0$ is given in figure 8.1. In figure 8.2, $\chi_{\perp}^H(\omega, T)$ as a function of ω for various values of T is shown. The numerous features of $\chi_{\perp}^H(\omega, T)$ mentioned above are manifest in these illustrations.

B. The Plane Square Lattice

An expression for the frequency dependent initial perpendicular susceptibility of the plane square lattice can be found in just the same

Fig 8.1.

The temperature dependence of the initial perpendicular susceptibility of the quantum mechanical Ising model on the honeycomb lattice for

a) $\omega = 0$ b) $\omega = 0.6\omega_0$ and c) $\omega = 0.8\omega_0$.

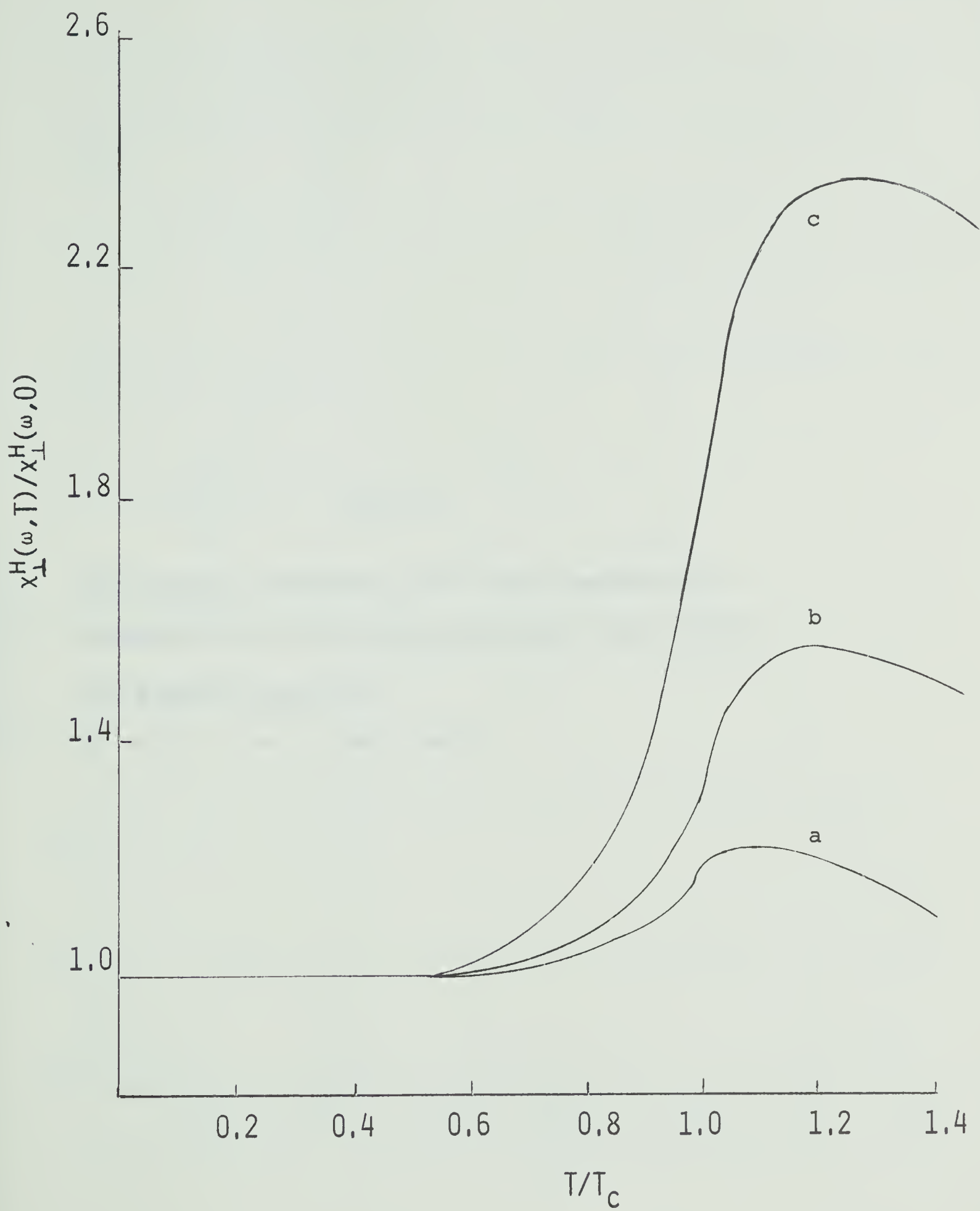
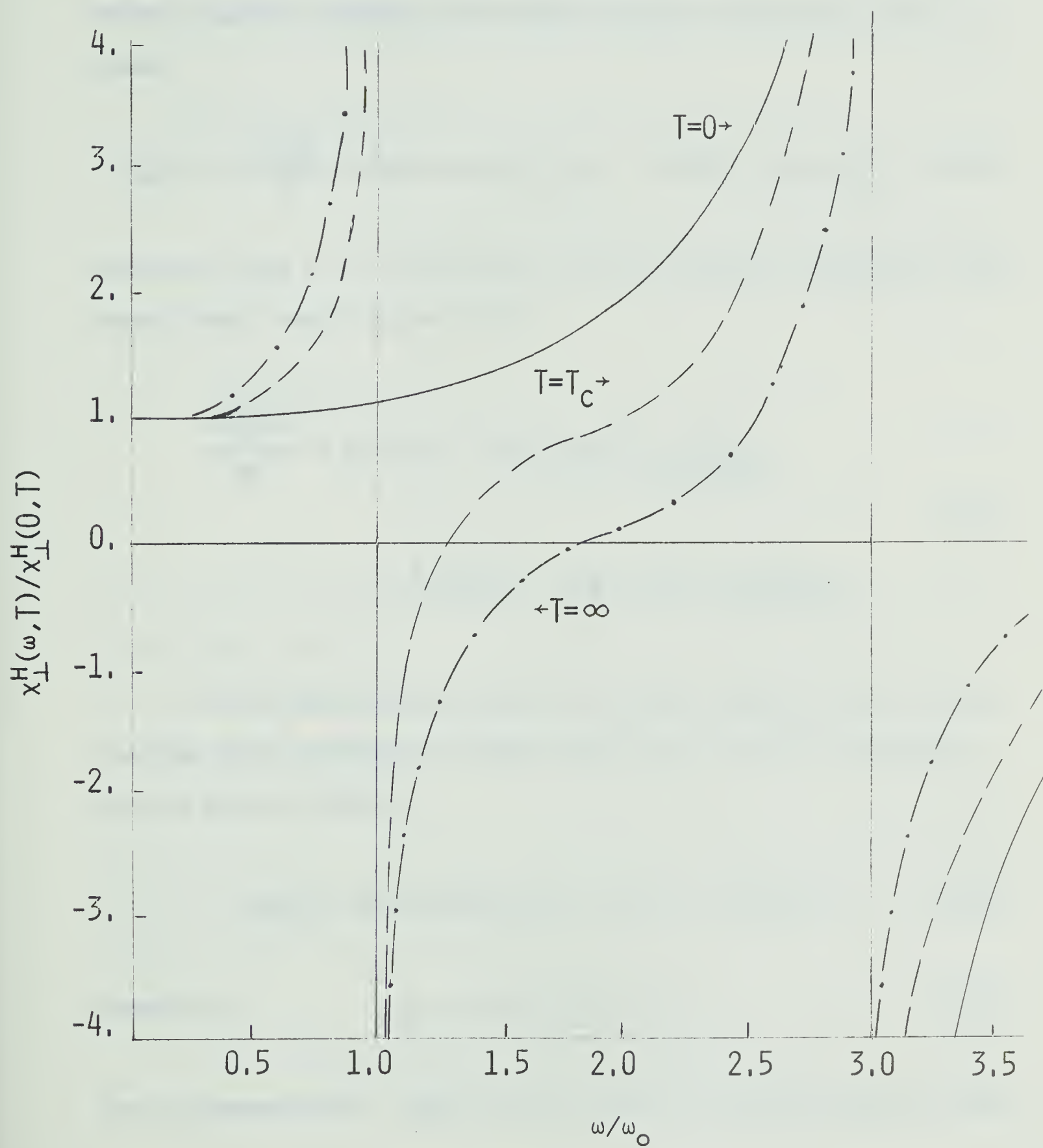


Fig 8.2

The frequency dependence of the initial perpendicular susceptibility of the quantum mechanical Ising model on the honeycomb lattice for

$T = 0$, $T = T_c$ and $T \rightarrow \infty$.



way as for the honeycomb lattice. For any regular $q = 4$ lattice (plane square, Kagome, diagonal), the response function expression (7.57) becomes

$$\phi_{xx}^s(t, T) = \frac{8Nm^2}{\hbar} \cos^3 \omega_0 t \sin \omega_0 t \left\{ \langle \sigma_0 \sigma_1 \rangle - \tan^2 \omega_0 t \langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle \right\} \quad (8.24)$$

where now spins 1, 2, 3 are three of the four nearest neighbours to spin zero. Again from (7.15) we obtain

$$\begin{aligned} \frac{J\chi_{\perp}^s(\omega, T)}{Nm^2} &= \frac{1}{8} [\langle \sigma_0 \sigma_1 \rangle + \langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle] \frac{1}{1 - (\omega/4\omega_0)^2} \\ &+ \frac{1}{2} [\langle \sigma_0 \sigma_1 \rangle - \langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle] \frac{1}{1 - (\omega/2\omega_0)^2} . \end{aligned} \quad (8.25)$$

For the plane square lattice the nearest neighbour pair correlation was first evaluated by Onsager (1944) and is given in convenient form by Fisher (1963) as,

$$\langle \sigma_0 \sigma_1 \rangle = \frac{1}{2} \coth 2K \left[1 + \frac{2}{\pi} (2 \tanh^2 2K - 1) \mathcal{K}(k_1) \right] , \quad (8.26)$$

$$\text{where now,} \quad k_1 = 2 \tanh 2K / \cosh 2K , \quad (8.27)$$

for all temperatures. Again the four particle correlation may be calcu-

lated by the transformation theorem described in the last section. By an exactly similar procedure we obtain first

$$\langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle = \frac{1}{8} (\tanh 4K - 2 \tanh 2K) + \frac{1}{2} \tanh 4K \langle \sigma_1 \sigma_2 \rangle \quad (8.28)$$

$$+ \frac{1}{4} \tanh 4K \langle \sigma_1 \sigma_3 \rangle + \frac{1}{8} (\tanh 4K + 2 \tanh 2K) \langle \sigma_1 \sigma_2 \sigma_3 \sigma_4 \rangle,$$

and also,

$$\langle \sigma_1 \sigma_2 \sigma_3 \sigma_4 \rangle = 1 + 2 \coth^2 2K - 4 \coth 2K (1 + \coth^2 2K) \times \quad (8.29)$$

$$\langle \sigma_0 \sigma_1 \rangle + 2 \coth^2 2K [2 \langle \sigma_1 \sigma_2 \rangle + \langle \sigma_1 \sigma_3 \rangle],$$

where $\langle \sigma_1 \sigma_2 \rangle$ and $\langle \sigma_1 \sigma_3 \rangle$ represent the second and third neighbour Ising correlations respectively. Then substituting (8.29) in (8.28) we get

$$\langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle = \coth 2K [2 \langle \sigma_1 \sigma_2 \rangle + \langle \sigma_1 \sigma_3 \rangle + 1] - (1 + 2 \coth^2 2K) \langle \sigma_0 \sigma_1 \rangle. \quad (8.30)$$

The second and third neighbour correlations, first worked out by Kaufman and Onsager (1949), are also given in a convenient form by Fisher (1963) which is valid for all temperatures i.e.

$$\langle \sigma_1 \sigma_2 \rangle = \frac{1}{\pi} \coth^2 2K [E(k_1) + k_1'' \mathcal{K}(k_1)] \quad (8.31)$$

and

$$\langle \sigma_1 \sigma_3 \rangle = \frac{1}{2} \coth^2 2K - \left(\frac{2}{\pi k_1} \right)^2 [E^2(k_1) - 2k_1'' \mathcal{K}(k_1) E(k_1) + (k_1'')^3 \mathcal{K}^2(k_1)] , \quad (8.32)$$

where k_1 is given in (8.27) and

$$k_1'' = 2 \tanh^2 2K - 1 . \quad (8.33)$$

$E(k_1)$ is the complete elliptic integral of the second kind defined by Jahnke and Emde (1945) as,

$$E(k_1) = \int_0^{\pi/2} \sqrt{1 - k_1^2 \sin^2 x} \, dx . \quad (8.34)$$

Equations (8.25) to (8.34) constitute an exact known solution to the perpendicular susceptibility of the plane square lattice. In many respects it is qualitatively very similar to that of the honeycomb lattice. There are resonances at $\omega = 4J/\hbar$ and $8J/\hbar$, just as we might expect, corresponding respectively to configurations in which three neighbours to a given spin are parallel and one antiparallel, and all four spins parallel. (We shall turn later to the case of two spins pointing in each direction.) Again the resonances have zero linewidth.

At $T = 0$, the resonance at $2\omega_0$ disappears as expected, and only the resonance at $4\omega_0$ corresponding to all spins parallel persists. As

$T \rightarrow \infty$ the amplitudes of the $2\omega_0$ and $4\omega_0$ resonances approach the expected ratio of 4 to 1. Each of the above remarks applies, of course, to any $q = 4$ lattice.

Near the critical temperature, given for the plane square lattice by $\sinh^2 2K_c^s = 1$, we again perform Taylor series expansions about T_c and obtain from (8.26), (8.31) and (8.32)

$$\langle \sigma_0 \sigma_1 \rangle = \frac{1}{\sqrt{2}} - \frac{4}{\pi} (K - K_c^s) \log |K - K_c^s|, \quad (8.35)$$

$$\langle \sigma_1 \sigma_2 \rangle = \frac{2}{\pi} - \frac{4\sqrt{2}}{\pi} (K - K_c^s) \log |K - K_c^s|, \quad (8.36)$$

and

$$\langle \sigma_1 \sigma_3 \rangle = \left(1 - \frac{4}{\pi^2}\right) - \frac{16\sqrt{2}}{\pi^2} (K - K_c^s) \log |K - K_c^s|. \quad (8.37)$$

Substituting in (8.30) gives

$$\langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle = \sqrt{2} \left[\frac{4(\pi - 1)}{\pi^2} - \frac{1}{2} \right] - \frac{4}{\pi^2} (8 - \pi) (K - K_c^s) \log |K - K_c^s|, \quad (8.38)$$

and hence for the analogue of (8.23) we get

$$\frac{J}{Nm^2} [\chi_{\perp}^s(\omega, T) - \chi_{\perp}^s(\omega, T_c^s)] = \frac{2K_c^s}{\pi^2} \left[\frac{4 - \pi}{1 - (\omega/2\omega_0)^2} - \frac{1}{1 - (\omega/4\omega_0)^2} \right] \left(1 - \frac{T}{T_c^s}\right) \log \left|1 - \frac{T}{T_c^s}\right|. \quad (8.39)$$

We see that again the perpendicular susceptibility is finite at T_c and has infinite slope. However, there is now the important difference that for low frequencies, the slope is negative infinite.

This provides only a premonition of the discrepancies to follow. If we now put $\omega = 0$ in (8.25) and substitute (8.30), we obtain

$$\frac{8J\chi_{\perp}^s(0,T)}{Nm^2} = 2(4 + 3 \coth^2 2K) \langle \sigma_0 \sigma_1 \rangle - 3 \coth 2K [2 \langle \sigma_1 \sigma_2 \rangle + \langle \sigma_1 \sigma_3 \rangle + 1] \quad (8.40)$$

whereas Fisher's static perpendicular susceptibility result is

$$\begin{aligned} \frac{8J\chi_{\perp}^s(T)}{Nm^2} = K \left\{ 6(1 + \coth^2 2K) - 12 \coth 2K(1 + \coth^2 2K) \langle \sigma_0 \sigma_1 \rangle \right. \\ \left. + 2(3 \coth^2 2K - 1)(2 \langle \sigma_1 \sigma_2 \rangle + \langle \sigma_1 \sigma_3 \rangle) \right\} + \frac{8J\chi_{\perp}^s(0,T)}{Nm^2} \end{aligned} \quad (8.41)$$

where in (8.41) $\chi_{\perp}^s(0,T)$ is given by (8.40). In other words Fisher's static result for the square lattice consists of our result for zero frequency plus an extra term containing a factor K . This is in striking contrast to the case for the honeycomb lattice where the zero frequency result and Fisher's static result agree exactly. The resolution of this discrepancy will be expounded both physically and mathematically in the next chapter.

CHAPTER IX

THE ZERO FREQUENCY LIMIT

We must now tackle the question of why our zero frequency results agree with Fisher's static results for the honeycomb lattice and not for the plane square lattice. More generally it seems that a static calculation of the type carried out by Fisher (1963) would agree with the $\omega \rightarrow 0$ dynamic result for any regular lattice of odd q and disagree for any regular lattice of even q . Indeed an examination of his equation (4.10) reveals how terms involving a factor K can arise for even q and not odd, whereas the Kubo formalism as applied in previous chapters seems unable to yield such factors for any lattice.

First we must realize, as Kubo (1957) himself emphasizes, that we have just calculated the susceptibility of an isolated system on which the magnetic field H_x has been imposed adiabatically (because of the $e^{\delta t}$ factor). On the other hand, Fisher's calculation is of the isothermal susceptibility of a system in thermal contact with a heat reservoir. Thus they need not be the same.

To see how this distinction might be important for even q lattices and not for odd q , we note that the former will contain at all non-zero temperatures configurations in which a central spin has half of its neighbours aligned parallel to it and the other half aligned antiparallel. Such a 'sublattice' of spins are effectively free spins.

It is well known that for a system of free spins there exists a great difference between the adiabatic and isothermal susceptibilities. In the former case, a free spin merely executes a Larmor precession about the direction of the applied field with no change in the component of the magnetic moment of the spin along the direction of the field. The adiabatic susceptibility is simply zero and it is no surprise then that our result (8.25) for the square lattice contains no contribution associated with such free spins. On the other hand, in a calculation of the isothermal susceptibility, it is always implicitly assumed that there exist weak interactions with the heat bath that are important in providing a mechanism by means of which the system can approach its equilibrium state in the presence of the magnetic field but that can be neglected in determining the nature of that equilibrium state. The isothermal susceptibility of a system of free spins is non-zero because of this assumption.

Our conclusion, then, is that the term $\chi_0^s(T)$ given by,

$$\begin{aligned} \frac{8J\chi_0^s(T)}{Nm^2} = K \left\{ 6(1 + \coth^2 2K) - 12 \coth 2K(1 + \coth^2 2K) \langle \sigma_0 \sigma_1 \rangle \right. \\ \left. + 2(3 \coth^2 2K - 1) (2 \langle \sigma_1 \sigma_2 \rangle + \langle \sigma_1 \sigma_3 \rangle) \right\} , \end{aligned} \quad (9.1)$$

which represents the difference between Fisher's result and ours can be considered as the contribution of the sublattice of free spins to the

isothermal initial perpendicular susceptibility of the Ising model on the plane square lattice. The contribution (9.1) appears because of the weak interactions discussed above and, as expected, does not appear in a calculation of the adiabatic perpendicular susceptibility, where such interactions are explicitly omitted.

To illustrate further the validity of this assertion, we shall introduce a mechanism by means of which it is possible to calculate $\chi_0^S(T)$ by the Kubo formalism. We now consider the problem where \mathcal{H}_0 is given not by (7.6) but by

$$\mathcal{H}_0 = J \sum_{nn} \sigma_i^z \sigma_j^z - mH_z \sum_i \sigma_i^z, \quad (9.2)$$

i.e. an external static magnetic field is introduced in the z direction. The perpendicular susceptibility can also be calculated with (9.2) as the unperturbed Hamiltonian. We can see that the formerly free spins can now be expected to give a contribution to the susceptibility as determined by the Kubo formalism and in particular, if the $\omega \rightarrow 0$ limit is taken first and then the $H_z \rightarrow 0$ limit, (essentially the reverse of what we did in chapter eight) we should be able to make a direct comparison with Fisher's result. By so ordering our limits we are doing precisely what is required, namely, considering an interaction which is important in guiding the system to its equilibrium state but unimportant in determining its details (such details depending primarily on the Ising

interaction.)

The calculation is very similar to that outlined in previous chapters and we shall only emphasize the important details here. Corresponding to the identity (7.34), we make use of the relationship,

$$e^{\pm \frac{it\mathcal{H}_0}{\hbar}} = \prod_{nn} \left[\cos\left(\frac{\omega_0 t}{2}\right) \pm i\sigma_i^z \sigma_j^z \sin\left(\frac{\omega_0 t}{2}\right) \right] \prod_j \left[\cos\left(\frac{\delta\omega t}{2}\right) \pm i\sigma_j^z \sin\left(\frac{\delta\omega t}{2}\right) \right], \quad (9.3)$$

where \mathcal{H}_0 is given by (9.2), $\delta\omega = 2mH_z/\hbar$ and the second product is over all lattice sites. Corresponding to (7.38), we get

$$\phi_{xx}(t, T, H_z) = \frac{im^2}{\hbar} (\cos \omega_0 t)^q \cos \delta\omega t \times \quad (9.4)$$

$$\sum_{i', j'} \left\langle \left[\prod_{k=1}^q (1 - i\sigma_k^z \sigma_{j'}^z, u), \sigma_{j'}^x \right] (1 - i\sigma_{i'}^z, u_1) \sigma_{i'}^x, \right. \\ \left. + \prod_{k=1}^q (1 - i\sigma_k^z \sigma_{i'}^z, u) [(1 - i\sigma_{i'}^z, u_1) \sigma_{i'}^x, \sigma_{j'}^x] \right\rangle$$

where $u_1 = \tan \delta\omega t$. For the plane square lattice, using the same procedures as before, we then obtain

$$\phi_{xx}(t, T, H_z) = \frac{8Nm^2}{\hbar} \cos(\delta\omega t) \left\{ \cos^3 \omega_0 t \sin \omega_0 t \langle \sigma_0 \sigma_1 \rangle_H \right.$$

$$\begin{aligned}
& - \cos \omega_0 t \sin^3 \omega_0 t \langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle_H \} + \frac{2Nm^2}{h} \sin \delta \omega t \times \\
& \left\{ \cos^4 \omega_0 t \langle \sigma_0 \rangle_H - \sin^2 \omega_0 t [4 \langle \sigma_0 \sigma_1 \sigma_2 \rangle_H + 2 \langle \sigma_0 \sigma_1 \sigma_3 \rangle_H] \right. \\
& \left. + \sin^4 \omega_0 t \langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \sigma_4 \rangle_H \right\} \quad (9.5)
\end{aligned}$$

where the subscript H indicates the presence of H_z and, as before, $\sigma = \sigma^z$. The term in (9.5) proportional to $\cos(\delta \omega t)$ corresponds to (8.24). When the fourier transform is taken to obtain its contribution to the perpendicular susceptibility, the zero frequency and zero field result (8.40) is obtained no matter in which order $\omega \rightarrow 0$ and $H_z \rightarrow 0$ limits are taken.

Simple trigonometry shows that the remainder of (9.5) separates into parts proportional to $\sin \delta \omega t$, $\sin \delta \omega t \cos 2\omega_0 t$ and $\sin \delta \omega t \cos 4\omega_0 t$. The first is given by

$$\frac{Nm^2}{4h} \sin \delta \omega t [3 \langle \sigma_0 \rangle_H - (4 \langle \sigma_0 \sigma_1 \sigma_2 \rangle_H + 2 \langle \sigma_0 \sigma_1 \sigma_3 \rangle_H) + 3 \langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \sigma_4 \rangle_H] \quad (9.6)$$

It can easily be demonstrated that the other two terms give no contribution to the zero field, zero frequency susceptibility. However the term (9.6) requires careful attention. Using (7.15), this gives the following contribution to the perpendicular susceptibility,

$$\chi_0^s(\omega, T, H_z) = \frac{Nm^2}{4\hbar} \frac{\delta\omega}{(\delta\omega)^2 - \omega^2} [3\langle\sigma_0\rangle_H - \quad (9.7)$$

$$(4\langle\sigma_0\sigma_1\sigma_2\rangle_H + 2\langle\sigma_0\sigma_1\sigma_3\rangle_H) + 3\langle\sigma_0\sigma_1\sigma_2\sigma_3\sigma_4\rangle_H] .$$

We see that this term can be associated with the formerly free spins where now an energy $\hbar\delta\omega$ is required to overturn one of them.

We now apply Fisher's transformation theorem with, however, a number of important differences from the procedure previously described. Corresponding to (8.8) we now have

$$T_0(\sigma_1, \sigma_2, \sigma_3, \sigma_4) = \text{trace}_0 [\sigma_0 e^{K(\sigma_0\sigma_1 + \sigma_0\sigma_2 + \sigma_0\sigma_3 + \sigma_0\sigma_4) + \alpha\sigma_0}] \quad (9.8)$$

where $\alpha = mH_z/kT$. However because of the $\alpha\sigma_0$ term in (9.8), it is no longer antisymmetric when all four nearest neighbour spins are simultaneously reversed. We must therefore write (8.9) in the form,

$$T_0(\sigma_1, \sigma_2, \sigma_3, \sigma_4) = \left\{ \epsilon + \alpha \sum_{r=1}^4 \sigma_r + \beta \sum_{t=1}^6 \sigma_{a_t} \sigma_{b_t} + \gamma \sum_{u=1}^4 \sigma_{a_u} \sigma_{b_u} \sigma_{c_u} \right. \\ \left. + \delta \sigma_1 \sigma_2 \sigma_3 \sigma_4 \right\} \text{trace}_0 e^{K(\sigma_0\sigma_1 + \sigma_0\sigma_2 + \sigma_0\sigma_3 + \sigma_0\sigma_4) + \alpha\sigma_0} . \quad (9.9)$$

To determine α , β , γ , δ and ϵ , we obtain five independent equations by

identifying (9.8) and (9.9) when a) all four nearest neighbour spins are +1, b) all four -1, c) three +1 and one -1, d) three -1 and one +1, and e) two +1 and two -1. Solving these equations and then substituting in the obvious extension of (8.11) to obtain $\langle \sigma_0 \rangle_H$, $\langle \sigma_0 \sigma_1 \sigma_2 \rangle_H$, $\langle \sigma_0 \sigma_1 \sigma_3 \rangle_H$ and $\langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \sigma_4 \rangle_H$, we finally obtain

$$\begin{aligned}
 & 3\langle \sigma_0 \rangle_H - 4\langle \sigma_0 \sigma_1 \sigma_2 \rangle_H - 2\langle \sigma_0 \sigma_1 \sigma_3 \rangle_H + 3\langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \sigma_4 \rangle_H \\
 & = \tanh \alpha [3 - 4\langle \sigma_1 \sigma_2 \rangle_H - 2\langle \sigma_1 \sigma_3 \rangle_H + 3\langle \sigma_1 \sigma_2 \sigma_3 \sigma_4 \rangle_H] .
 \end{aligned} \tag{9.10}$$

Inserting (9.10) in (9.7) and taking limits, we get

$$\begin{aligned}
 \lim_{H_z \rightarrow 0} \lim_{\omega \rightarrow 0} \chi_0^s(\omega, T, H_z) &= \frac{Nm^2}{4\pi} [3 - 2(2\langle \sigma_1 \sigma_2 \rangle + \langle \sigma_1 \sigma_3 \rangle) \\
 &+ 3\langle \sigma_1 \sigma_2 \sigma_3 \sigma_4 \rangle] \lim_{H_z \rightarrow 0} \frac{\tanh \frac{mH_z}{kT}}{\frac{2mH_z}{\hbar}} ,
 \end{aligned} \tag{9.11}$$

$$\text{i.e.} \quad \frac{8J\chi_0^s(0, t, 0)}{Nm^2} = K[3 - 2(2\langle \sigma_1 \sigma_2 \rangle + \langle \sigma_1 \sigma_3 \rangle) + 3\langle \sigma_1 \sigma_2 \sigma_3 \sigma_4 \rangle], \tag{9.12}$$

and the correlations in (9.11) and (9.12) are evaluated in zero field. Then substituting (8.29) in (9.12), we immediately obtain,

$$\chi_0^s(0,T,0) = \chi_0^s(T), \quad (9.13)$$

where $\chi_0^s(T)$ is given in (9.1).

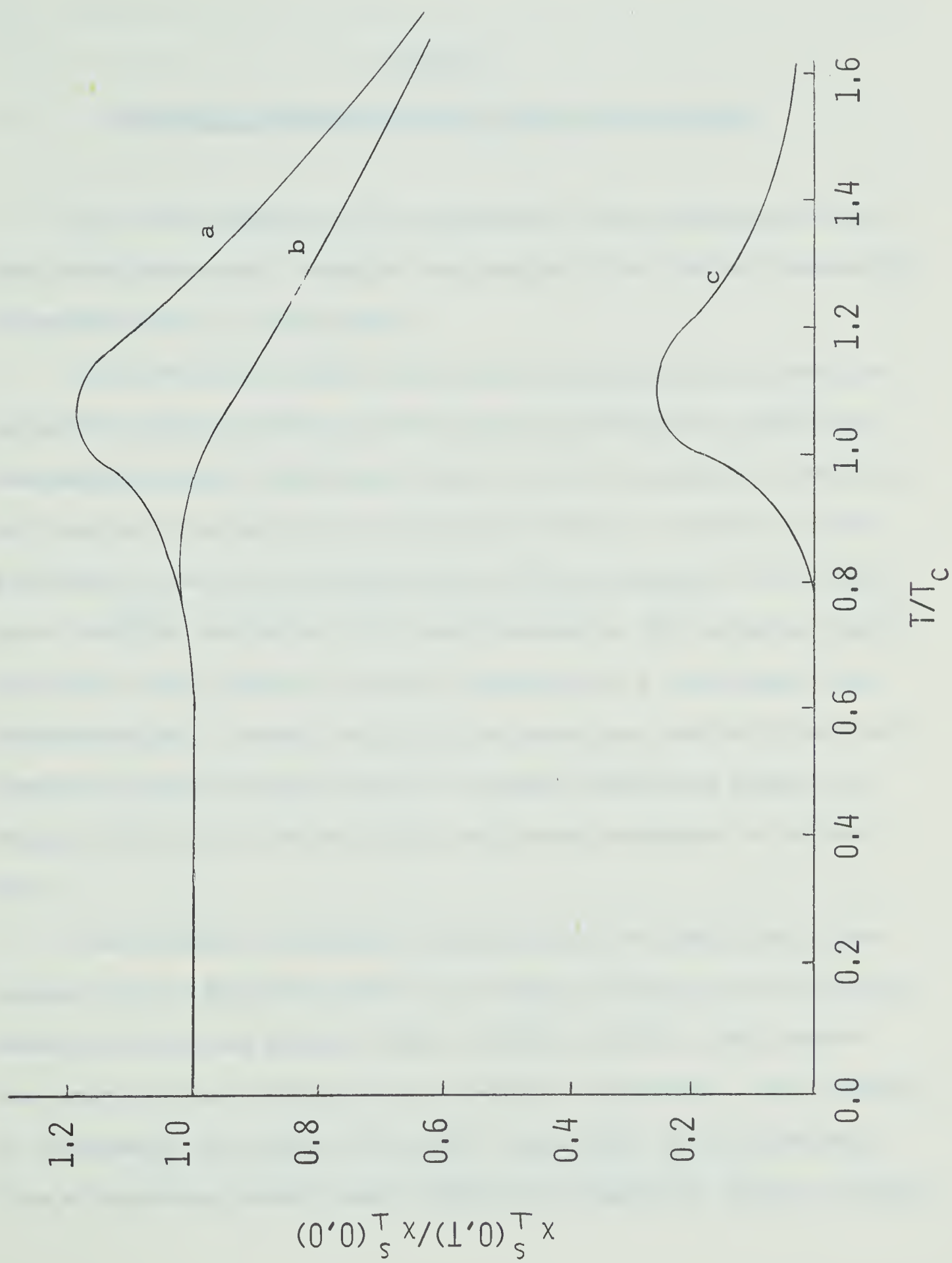
This result confirms our conclusions regarding the distinction between the adiabatic susceptibility calculated by the Kubo formalism and the isothermal susceptibility evaluated by Fisher for the plane square lattice. It is important to note that, for lattices of odd coordination number, one does not obtain terms proportional to $\sin \delta \omega t$ alone as in (9.5) and hence no additional contributions will be found by reversing the limits.

In Figure 9.1 we plot for the plane square lattice the isothermal susceptibility given by Fisher's (1963) equation (5.42), the zero frequency adiabatic susceptibility in our equation (8.25) together with their difference $\chi_0^s(T)$. We see that the difference is appreciable only in the critical region. Certainly that the two susceptibilities are equal at zero temperature is consistent with the non-existence in the completely magnetized state of any 'free' spins.

Fig. 9.1

The temperature dependence of the zero frequency initial perpendicular susceptibility of the quantum mechanical Ising model on the square lattice

- (a) the isothermal susceptibility, $\chi_{\perp}^s(T)$
- (b) the adiabatic susceptibility $\chi_{\perp}^s(0,T)$ and
- (c) the difference between the two susceptibilities.



CHAPTER X

INELASTIC NEUTRON SCATTERING FROM AN ISING MAGNET

As a final example of the application of the techniques we have just described we shall consider the problem of the inelastic scattering of neutrons from an Ising system.

Since Van Hove (1954) first point out and explicitly stated the connection between neutron scattering from ferromagnetic crystals and time-dependent spin correlations, there has been considerable theoretical interest in establishing microscopic theories of neutron magnetic scattering. One of the primary aims has been to describe the phenomenon of critical scattering, the large increase in the scattering amplitude that occurs near the critical temperature of a ferromagnet (and antiferromagnet). Notable contributions have been made by Elliott and Marshall (1958) and more recently by Marshall (1965) and Fisher and Burford (1967) all of which contain many other references to relevant work.

The problem of inelastic scattering has been specifically considered for the Heisenberg model by De Gennes (1958) and for the Ising model by Collins and Windsor (1967) and Sears (1967a). Both papers deal only with the limiting case of infinite temperature. Very recently, by considering the effect of the short range order on the scattering from a Heisenberg magnet, Sears (1967b) has extended De Gennes's original

work. However, it has not been possible thus far to make accurate predictions regarding the critical scattering from a Heisenberg system where fluctuations in the long range order are important. In contrast, we shall demonstrate that in the much simpler Ising model, it is possible to determine the exact energy distribution of the inelastic scattering for the particular case of two dimensional spin one half systems at all temperatures.

For a system of N spins rigidly fixed to lattice sites, the cross section for the scattering of neutrons per unit solid angle Ω per unit energy E is given by (Van Hove 1954),

$$\frac{d^2\sigma}{d\Omega dE} = \left(\frac{\gamma e^2}{mc^2}\right)^2 \frac{N}{\hbar} \frac{k_1}{k_0} \left|f(\underline{K}')\right|^2 \sum_{\alpha\beta} (\delta_{\alpha\beta} - \hat{K}_\alpha \hat{K}_\beta) S^{\alpha\beta}(\underline{K}', \omega), \quad (10.1)$$

where e and m are the electron charge and mass, $\gamma = 1.91$ is the neutron magnetic moment in nuclear magnetons, c is the velocity of light, k_0 and $k_1 = k_0 - \underline{K}'$ are the initial and final wave vectors of the neutron, $\hbar\omega = \hbar(k_0^2 - k_1^2)/2m_0$, m_0 being the mass of the neutron, is the energy transferred from neutron to spin system and \hat{K}_α is the α th direction cosine of \underline{K}' . $f(\underline{K}')$ is the usual atomic form factor, the fourier transform of the electron spin density, and $S^{\alpha\beta}(\underline{K}', \omega)$ is given by,

$$S^{\alpha\beta}(\underline{K}', \omega) = \frac{1}{2\pi} \sum_{\underline{R}} \int_{-\infty}^{+\infty} \exp[i(\underline{K}' \cdot \underline{R} - \omega t)] \langle \sigma_{\underline{0}}^\alpha(0) \sigma_{\underline{R}}^\beta(t) \rangle dt. \quad (10.2)$$

$\sigma_{\underline{R}}^{\beta}(t)$, the β component of the spin on lattice site \underline{R} at time t is of the form,

$$\sigma_{\underline{R}}^{\beta}(t) = \exp(i\mathcal{H}_0 t/\hbar) \sigma_{\underline{R}}^{\beta} \exp(-i\mathcal{H}_0 t/\hbar) \quad , \quad (10.3)$$

with \mathcal{H}_0 given by (7.6).

As pointed out by Collins and Windsor (1967), for the Ising model $S^{\alpha\beta}(\underline{K}', \omega) = -S^{\beta\alpha}(\underline{K}', \omega)$ for $\alpha \neq \beta$ and so the "off diagonal" elements make no contribution to the scattering cross section (10.1). Simple inspection also shows that $S^{zz}(\underline{K}', \omega)$ is responsible for the critical elastic scattering and has been very completely investigated by Fisher and Burford (1967). In contrast to the Heisenberg model, the inelastic scattering is solely determined by $S^{xx}(\underline{K}', \omega) = S^{yy}(\underline{K}', \omega)$ which is essentially what Collins and Windsor calculated in the infinite temperature limit. We shall proceed to determine $S^{xx}(\underline{K}', \omega)$ exactly for all temperatures by techniques very similar to those used in previous chapters.

Substituting (7.34) in (10.3) we obtain, after some manipulation,

$$\left\langle \sigma_{\underline{0}}^x \sigma_{\underline{R}}^x(t) \right\rangle = \cos^4 \omega_0 t \left\langle \sigma_{\underline{0}}^x \sigma_{\underline{R}}^x \prod_{k=1}^4 (1 + i \sigma_{\underline{k}}^z \sigma_{\underline{R}}^z \tan \omega_0 t) \right\rangle \quad (10.4)$$

where, as before, $\omega_0 = 2J/\hbar$. (10.4) and all future remarks unless otherwise stated refer to the particular case of the plane square lattice (or any lattice for which $q = 4$). However, an exactly similar analysis could be undertaken for any known lattice. It is clear that

for the Ising model as given in (7.6) non zero correlations can exist only between the z components of spins on different lattice sites. Hence (10.4) is non zero only for $\underline{R} = 0$ and on expanding the right hand side, we obtain

$$\begin{aligned} \langle \sigma_0^x \sigma_0^x(t) \rangle &= \cos^4 \omega_0 t + 4i \cos^3 \omega_0 t \sin \omega_0 t \langle \sigma_0 \sigma_1 \rangle \\ &- 2 \cos^2 \omega_0 t \sin^2 \omega_0 t [2 \langle \sigma_1 \sigma_2 \rangle + \langle \sigma_1 \sigma_3 \rangle] \\ &- 4i \cos \omega_0 t \sin^3 \omega_0 t \langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle + \sin^4 \omega_0 t \langle \sigma_1 \sigma_2 \sigma_3 \sigma_4 \rangle \end{aligned} \quad (10.5)$$

where as before we have dropped the superscript z from the spins on the right hand side and 1, 2, 3, 4 label the nearest neighbour sites to the zeroth. Substitution of (10.5) in (10.2) then gives

$$\begin{aligned} S^{xx}(\underline{k}', \omega, T) &= \delta(\omega) \left[\frac{3}{8} - \frac{1}{4} (2 \langle \sigma_1 \sigma_2 \rangle + \langle \sigma_1 \sigma_3 \rangle) + \frac{3}{8} \langle \sigma_1 \sigma_2 \sigma_3 \sigma_4 \rangle \right] \\ &+ \frac{1}{2} \delta(\omega - 2\omega_0) \left[\frac{1}{2} + \langle \sigma_0 \sigma_1 \rangle - \frac{1}{2} \langle \sigma_1 \sigma_2 \sigma_3 \sigma_4 \rangle - \langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle \right] \\ &+ \frac{1}{2} \delta(\omega - 4\omega_0) \left[\frac{1}{8} + \frac{1}{2} \langle \sigma_0 \sigma_1 \rangle + \frac{1}{4} (2 \langle \sigma_1 \sigma_2 \rangle + \langle \sigma_1 \sigma_3 \rangle) \right. \\ &\left. + \frac{1}{2} \langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle + \frac{1}{8} \langle \sigma_1 \sigma_2 \sigma_3 \sigma_4 \rangle \right] + \frac{1}{2} \delta(\omega + 2\omega_0) \left[\frac{1}{2} - \langle \sigma_0 \sigma_1 \rangle \right. \\ &\left. - \frac{1}{2} \langle \sigma_1 \sigma_2 \sigma_3 \sigma_4 \rangle + \langle \sigma_0 \sigma_1 \sigma_2 \sigma_3 \rangle \right] + \frac{1}{2} \delta(\omega + 4\omega_0) \left[\frac{1}{8} - \frac{1}{2} \langle \sigma_0 \sigma_1 \rangle \right] \end{aligned}$$

$$+\frac{1}{4}(2\langle\sigma_1\sigma_2\rangle+\langle\sigma_1\sigma_3\rangle)+\frac{1}{8}\langle\sigma_1\sigma_2\sigma_3\sigma_4\rangle-\frac{1}{2}\langle\sigma_0\sigma_1\sigma_2\sigma_3\rangle] \quad (10.6)$$

The temperature dependence of all the spin correlations in (10.6) for the plane square lattice have been discussed in chapter eight and (10.6) then constitutes an exact solution of both the temperature dependence and energy distribution of the inelastic scattering of neutrons from the plane square spin one half Ising lattice.

In contrast to the Heisenberg model, the energy distribution consists of a number of discrete levels, each level corresponding to a particular orientation of the four nearest neighbours to any given spin. For instance, as before, the $\delta(\omega - 4\omega_0)$ contribution is associated with the configuration in which all four spins are parallel with the central one only now the four antiparallel neighbouring spin environment makes a physically meaningful contribution to the $\delta(\omega + 4\omega_0)$ peak. The former represents absorption of energy by the spins, the latter by the neutrons.

The temperature dependent amplitudes of these levels given in (10.6) in terms of spin correlation functions are proportional to the probability that the associated spin orientation may occur. For instance, as we might expect by now, at $T = 0$ only the $\omega = + 4\omega_0$ level has non zero amplitude since in the completely magnetized state only the all parallel configuration will arise. Also the fact that the energy distribution is symmetric in only the infinite temperature limit is consistent with this physical interpretation. Indeed since all spin correlations tend to zero in this limit, we see by inspection of (10.6) that the $\omega = 0$,

$\omega = \pm 2\omega_0$ and $\omega = \pm 4\omega_0$ levels are in the ratio of 6 to 4 to 1. This is precisely the ratio of the probabilities of occurrence of the corresponding spin configurations in a random array of spins. From equations (8.35) to (8.38), we recall that all the spin correlations have infinite slope at the critical temperature and vary very little outside a relatively narrow region around T_c . This means that the energy distribution varies appreciably only in the critical region. Many of the above features are illustrated in Figure 10.1 where $S^{xx}(\underline{K}', \omega, T)$ is plotted versus ω for $T = 0$, $T = 0.8 T_c$, $T = T_c$, $T = 2 T_c$ and $T \rightarrow \infty$.

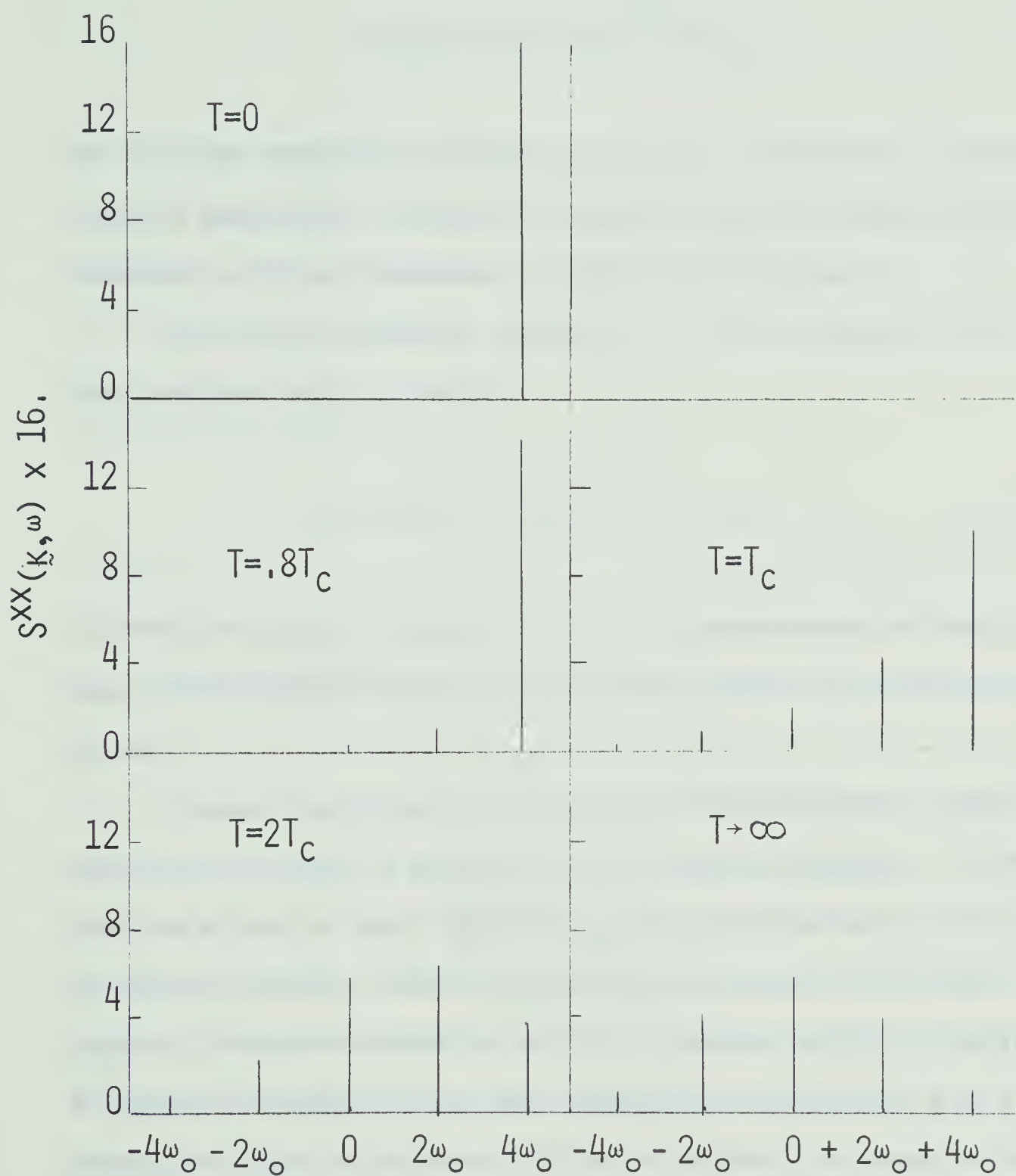
Much that we have said about the scattering from two dimensional Ising magnets and the plane square lattice in particular will also be characteristic of scattering by three dimensional Ising magnets. Firstly we should emphasize once again that an expression of the form (10.6) for $S^{xx}(\underline{K}', \omega, T)$ can be found without difficulty for a regular lattice of any coordination number and irrespective of dimension. For three dimensional lattices, however, the correlations cannot be found exactly but the high and low temperature series expansions for them could be calculated. Then the entire temperature dependence of each would be well represented by Padé approximant interpolation and extrapolation. We know further from the work of Sykes, Martin and Hunter (1967) that close to T_c for $T > T_c$,

$$C_H \underset{H \rightarrow 0}{\propto} (T - T_c)^{-1/8} . \quad (10.7)$$

This means that for the nearest neighbour correlation $\langle \sigma_0 \sigma_1 \rangle$ which is

Fig. 10.1

Inelastic neutron scattering amplitudes from a plane square Ising lattice for $T = 0$, $T = 0.8T_c$, $T = T_c$, and $T \rightarrow \infty$.



proportional to the energy,

$$\langle \sigma_0 \sigma_1 \rangle = C_1 (T - T_c)^{7/8} + C_2 \quad (10.8)$$

for the same temperature range (C_1 and C_2 are constants). The same type of rapid temperature variation is expected for all nearest neighbour correlations in three dimensions and hence for $S^{xx}(\underline{k}', \omega, T)$.

As in two dimensions, energy can be lost or gained by the scattered neutrons only in amounts

$$E = \pm \hbar \omega = 2qJ, (2qJ - 4)J, \dots \quad (10.9)$$

We therefore expect that qualitatively the temperature and energy dependence of the scattered neutrons will be the same in three dimensions as in two.

Finally the effect of interactions with neighbours farther than the first is simply to introduce further sharp resonances. On the other hand the effect of small $\sigma_i^x \sigma_j^x$ and $\sigma_i^y \sigma_j^y$ type interactions, to be expected in any real crystal, would be principally to round out the sharp resonances in energy distribution and also introduce slight \underline{k}' dependence in $S^{xx}(\underline{k}', \omega, T)$ because in that case correlations between the x or y components of spins on different lattice sites would no longer be zero.

CHAPTER XI

FINAL REVIEW AND DISCUSSION

We have considered in this thesis two problems whose many differences illustrate both the variety of lattice models of critical phenomena and the many distinct techniques that have been developed to investigate them.

A. Spin One Exchange Model

In the spin one exchange model, we have examined the equilibrium critical behavior of a model with a completely isotropic Hamiltonian. We have already noted in chapter one that short range interaction models (usually just nearest neighbours) give results which approximate very closely to experiment. Also these results are quite different from those obtained from the mean field calculations which are equivalent to systems with infinitely long range interactions. From the theoretical point of view, it seems clear that the range of the interaction is an important parameter controlling the macroscopic critical behavior of the system. The question still remains, however, of how relevant are the details of the interaction, i.e. whether it is pure Heisenberg or spin one exchange for instance, when the range is kept constant. This is largely the interest in our calculation on this model by means of which we hope to shed some light on this question.

Because of the inherent complexity of the calculation, it has not been possible to obtain an exact solution. The technique of high temperature series expansions is, however, a very precise means of obtaining successive approximations. The series in powers of inverse temperature represent an expansion about the infinite temperature limit where succeeding terms take into account the increasing order in the system as the temperature is lowered. The aim is to determine exactly as many of the coefficients of the various series as is reasonably possible. These truncated series in themselves fail to represent precisely the corresponding thermodynamic variable for temperatures very close to critical but extrapolation techniques such as the Domb ratio method and the Pade approximant method have succeeded in estimating the critical behavior with considerable precision.

There exist two methods of obtaining the series coefficients, the direct method and the linked cluster method, and we have shown how the latter is essentially a rearrangement of the former. Nevertheless, as we have emphasized, it is a much less complicated and more systematic technique to apply particularly with the further simplifications introduced by using the representation theory of the symmetric group. By this method we have obtained longer series than have previously been calculated for a quantum mechanical spin one system.

The numerical values we obtained for the critical temperature, the critical entropy and γ , the high temperature critical index of the magnetic susceptibility clearly indicate that there are small but sig-

nificant differences in critical behavior which can be directly attributed to the details of the nearest neighbour interaction. It would seem then that any theory based on the assumption that such details are not important (e.g. Kadanoff 1966) can only achieve qualitative and not quantitative consistency with the model calculations.

Using Domb and Miedema's (1964) and Heller's (1967) experimental reviews as our main references, we know of no spin one insulating ferromagnet whose experimental behavior might be compared to our results. This is not to say that such substances do not exist or may not be discovered. If such experiments were performed, we certainly would expect that some of the trends our results show in comparison with the spin one Heisenberg model would be manifest in some of these substances.

A recent controversy has surrounded the possibility of a non-zero critical temperature in the two-dimensional Heisenberg model. On the one hand, Mermin and Wagner (1966) have proved rigorously that a two-dimensional Heisenberg model cannot have a spontaneous magnetization or sublattice magnetization. On the other, Stanley and Kaplan (1966) have pointed out that the evidence from high-temperature series expansions for χ_0 for the Heisenberg model in two dimensions favours a non-zero critical temperature, especially for $S > \frac{1}{2}$. We have examined the high temperature series of the spin one exchange model for the triangular lattice, the two dimensional lattice with the highest coordination number and, therefore, presumably the most regular behavior. The results of the analysis, based on longer series than Stanley and Kaplan have

available for the Heisenberg model, are presented in appendix F. Our interpretation is that with the series available, there is no evidence for the existence of a critical temperature in two dimensional spin one exchange models.

There remains at least one interesting theoretical question about spin one systems. Just as there are two linearly independent isotropic interactions for spin one particles e.g. $\vec{S}_i \cdot \vec{S}_j$ and $(\vec{S}_i \cdot \vec{S}_j)^2$ so there are two independent long range order parameters suggested by these interactions. The Heisenberg interaction $\vec{S}_i \cdot \vec{S}_j$ between nearest neighbour pairs suggests the usual long-range order parameter,

$$\begin{aligned} R_1 &= \langle \vec{S}_0 \cdot \vec{S}_\infty \rangle = 3 \langle S_0^z S_\infty^z \rangle \\ &= 3 \frac{\bar{M}^2}{m^2} \end{aligned} \tag{11.1}$$

where the subscripts indicate the limit of infinite separation of the two spins and \bar{M} is the spontaneous magnetization per spin. The second long range order parameter may be taken to be $\langle (\vec{S}_0 \cdot \vec{S}_\infty)^2 \rangle$ but the preferable definition for both computational and theoretical reasons is

$$R_2 = \langle P_{0\infty} \rangle \tag{11.2}$$

where $P_{0\infty}$ is the exchange operator for two particles in the limit of

infinite separation. For spin one half systems the two definitions are of course linearly related.

In the high temperature region ($T > T_c$) all long range order is absent but the fluctuation in the long range order parameters is non-zero and becomes very large just above T_c . In fact the fluctuation ΔR_1 is directly proportional to the initial susceptibility which we have just determined. It would be interesting then to investigate ΔR_2 . Presumably ΔR_2 also has a singularity at T_c with some critical index γ_2 but it is not clear that it would be equal to γ for the spin one exchange model.

B. The Perpendicular Susceptibility of the Ising Model

In complete contrast, in this problem we have calculated a time-dependent macroscopic variable for a system whose nearest neighbour interaction is highly anisotropic. Indeed it is this anisotropy that is largely responsible for the time dependent behavior. Again, in contrast to the spin one exchange model, the Ising interaction is sufficiently elementary that we have been able to calculate the frequency dependent initial perpendicular susceptibility exactly for two dimensional systems.

The formalism we have used, the Kubo linear response theory, is particularly useful for studying the response of a system to a weak external, mechanical disturbance. The linear approximations embodied in the theory become exact in our case of the external field tending to

zero.

In comparing our results with Fisher's static calculations, we have emphasized the important distinction between the susceptibility of an isolated system and of one in contact with a heat bath or some agency that allows the system to reach its equilibrium state in the presence of the external magnetic field. In particular we found that for the simple quadratic lattice, and most likely all even coordination number lattices, there exists a quantitative difference which can be directly attributed to whether the system is isolated or not.

The point can also be made that in comparison with Glauber's (1963) master equation approach, our results contain no additional unknown parameters. Any time dependent variable of a system in contact with a heat bath will contain properties that depend not only on the basic interaction between the constituents of the system (e.g. the Ising interaction) but also on the nature of the external contact. This is reflected in the presence of unknown parameters in Glauber's results. However, our results both for the perpendicular susceptibility and the neutron scattering cross-section are wholly dependent on the nature of the Ising interaction. As a consequence, similar results say for the Heisenberg model will be completely different in nature and will provide a very good basis on which to decide whether a given system has or has not any anisotropy in its basic interaction. Indeed, the frequency dependent perpendicular susceptibility differentiates between such models to a much greater extent than do the static parallel susceptibilities.

Again the experimental data on the perpendicular susceptibility of Ising systems is very limited. Fisher (1965) points out some work performed by Lasheen et al. (1958) on the isothermal perpendicular susceptibility of the antiferromagnet $\text{MnCl}_2\text{H}_2\text{O}$. At zero frequency there is qualitative agreement only between his results (and therefore ours) and experiment. However, much of the discrepancy may be due to the fact that $\text{MnCl}_2\text{H}_2\text{O}$ inevitably only approximates to an Ising system.

It seems unlikely that exact results will ever be obtained both for the frequency dependent susceptibility and the neutron scattering cross section for three dimensional Ising or Heisenberg systems. However, it would seem a useful approach to apply the high temperature series expansion techniques we have described in Part I together with the Kubo formalism we introduced in Part II to make much more precise approximate calculations than have thus far been attempted.

The work we have just described in this thesis is also reported in a series of four papers, one of which deals with the spin one exchange model (Allan and Betts 1967), one with the inelastic neutron scattering (Allan and Betts 1968c) and two with the frequency dependent initial perpendicular susceptibility (Allan and Betts 1968a,b)).

We should also like to mention four recent papers by other authors which either make reference to the above publications or have arisen partly as a result of private communication between the authors and ourselves. Seagraves (1966) gives an alternative derivation of the spin representation of the permutation operator for spin $1/2$, 1 , $3/2$

and 2. Joseph (1967) has considered the problem of the exchange interaction for general spin in comparison with Heisenberg interaction. Falk (1968) demonstrates that the adiabatic susceptibility is a lower bound to the isothermal susceptibility and uses our calculation in part II of this thesis as an example. Finally Kim (1968) constructs a frequency dependent susceptibility which equals the static isothermal susceptibility in the zero frequency limit and agrees with our adiabatic susceptibility for non-zero frequencies.

APPENDIX AThe cumulant-moment identities for $\mu = 0$.

$$\lambda_1 = \mu_1 = 0$$

$$\lambda_2 = \mu_2$$

$$\lambda_3 = \mu_3$$

$$\lambda_4 = \mu_4 - 3\mu_2^2$$

$$\lambda_5 = \mu_5 - 10\mu_3\mu_2$$

$$\lambda_6 = \mu_6 - 15\mu_4\mu_2 - 10\mu_3^2 + 30\mu_2^3$$

$$\lambda_7 = \mu_7 - 21\mu_5\mu_2 - 35\mu_4\mu_3 + 210\mu_3\mu_2^2$$

$$\lambda_8 = \mu_8 - 28\mu_6\mu_2 - 56\mu_5\mu_3 - 35\mu_4^2 + 420\mu_4\mu_2^2 + 560\mu_3^2\mu_2 - 630\mu_2^4$$

$$\begin{aligned} \lambda_9 = \mu_9 - 36\mu_7\mu_2 - 84\mu_6\mu_3 - 126\mu_5\mu_4 + 756\mu_5\mu_2^2 + 2520\mu_4\mu_3\mu_2 + 560\mu_3^3 \\ - 7560\mu_3\mu_2^3 \end{aligned}$$

— | —

$$\lambda_2'' = \mu_2''$$

$$\lambda_3'' = \mu_3''$$

$$\lambda_4'' = \mu_4'' - 6\mu_2\mu_2''$$

$$\lambda_5'' = \mu_5'' - 10(\mu_3\mu_2'' + \mu_2\mu_3'')$$

$$\lambda_6'' = \mu_6'' - 15(\mu_4\mu_2'' + \mu_2\mu_4'') - 20\mu_3\mu_3'' + 90\mu_2^2\mu_2''$$

$$\lambda_7'' = \mu_7'' - 21(\mu_5\mu_2'' + \mu_2\mu_5'') - 35(\mu_4\mu_3'' + \mu_3\mu_4'') + 210(2\mu_3\mu_2\mu_2'' + \mu_2^2\mu_3'')$$

$$\begin{aligned} \lambda_8'' = \mu_8'' - 28(\mu_6\mu_2'' + \mu_2\mu_6'') - 56(\mu_5\mu_3'' + \mu_3\mu_5'') - 70\mu_4\mu_4'' + 420(\mu_4''\mu_2^2 \\ + 2\mu_2\mu_4\mu_2'') + 560(\mu_3^2\mu_2'' + 2\mu_3\mu_2\mu_3'') - 2520\mu_2^3\mu_2'' \end{aligned}$$

$$\begin{aligned} \lambda_9'' = \mu_9'' - 36(\mu_7\mu_2'' + \mu_2\mu_7'') - 84(\mu_6\mu_3'' + \mu_3\mu_6'') - 126(\mu_5\mu_4'' + \mu_4\mu_5'') \\ + 756(\mu_5''\mu_2^2 + 2\mu_5\mu_2\mu_2'') + 252(\mu_4''\mu_3\mu_2 + \mu_4\mu_2\mu_3'' + \mu_4\mu_3\mu_2'') \\ + 1680\mu_3^2\mu_2'' - 7560(\mu_3''\mu_2^3 + 3\mu_3\mu_2\mu_2'') \end{aligned}$$

APPENDIX BDecomposition of the Permutation Operator

$$P_{ij} - \frac{1}{3} = \sum_{n=1}^{12} a_i^n a_j^n$$

where

$$a_i^1 = (S_i^x)^2 - \frac{2}{3}$$

$$a_i^7 = S_i^y S_i^x$$

$$a_i^2 = (S_i^y)^2 - \frac{2}{3}$$

$$a_i^8 = S_i^z S_i^y$$

$$a_i^3 = (S_i^z)^2 - \frac{2}{3}$$

$$a_i^9 = S_i^x S_i^z$$

$$a_i^{14} = S_i^x S_i^y$$

$$a_i^{10} = S_i^x$$

$$a_i^5 = S_i^y S_i^z$$

$$a_i^{11} = S_i^y$$

$$a_i^6 = S_i^z S_i^x$$

$$a_i^{12} = S_i^z$$

APPENDIX C

Character Tables of the Irreducible Representations of $\Pi_2 \rightarrow \Pi_7$

Notation: Γ_n represents the 3^N dimensional reducible direct product representation of Π_N . Only non zero $n(\gamma)$ are listed.

Π_2

CLASS	1^2	2
ORDER	1	1
(2)	1	1
(1^2)	1	-1
Γ_2	9	3

$$n(2) = 6$$

$$n(1^2) = 3$$



Π_3

CLASS	1^3	$1,2$	3
ORDER	1	3	2
(3)	1	1	1
$(2,1)$	2	0	-1
(1^3)	1	-1	1
Γ_3	27	9	3

$$n(3) = 10$$

$$n(1^3) = 1$$

$$n(2,1) = 8$$

————— | —————

 Π_4

CLASS	1^4	$1^2,2$	$1,3$	4	2^2
ORDER	1	6	8	6	3
(4)	1	1	1	1	1
$(3,1)$	3	1	0	-1	-1
(2^2)	2	0	-1	0	2
$(2,1^2)$	3	-1	0	1	-1
(1^4)	1	-1	1	-1	1
Γ_4	81	27	9	3	9

$$n(4) = 15$$

$$n(2^2) = 6$$

$$n(3,1) = 15$$

$$n(2,1^2) = 3$$

Π_5	CLASS	1^5	$1^3,2$	$1^2,3$	$1,4$	$1,2^2$	$2,3$	5
	ORDER	1	10	20	30	15	20	24
	(5)	1	1	1	1	1	1	1
	(4,1)	4	2	1	0	0	-1	-1
	(3,2)	5	1	-1	-1	1	1	0
	(3,1 ²)	6	0	0	0	-2	0	1
	(2 ² ,1)	5	-1	-1	1	1	-1	0
	(2,1 ³)	4	-2	1	0	0	1	-1
	(1 ⁵)	1	-1	1	-1	1	-1	1
	\square_5	243	81	27	9	27	9	3

$$n(5) = 21$$

$$n(4,1) = 24$$

$$n(2^2,1) = 3$$

$$n(3,2) = 15$$

$$n(3,1^2) = 6$$



Π_6

CLASS	1^6	$1^4,2$	$1^3,3$	$1^2,4$	$1^2,2^2$	$1,2,3$	$1,5$	6	$2,4$	2^3	3^2
ORDER	1	15	40	90	45	120	144	120	90	15	4
(6)	1	1	1	1	1	1	1	1	1	1	1
(5,1)	5	3	2	1	1	0	0	-1	-1	-1	-1
(4,2)	9	3	0	-1	1	0	-1	0	1	3	0
(4,1 ²)	10	2	1	0	-2	-1	0	1	0	-2	1
(3 ²)	5	1	-1	-1	1	1	0	0	-1	-3	2
(3,2,1)	16	0	-2	0	0	0	1	0	0	0	-2
(2 ³)	5	-1	-1	1	1	-1	0	0	-1	3	2
(3,1 ³)	10	-2	1	0	-2	1	0	-1	0	2	1
(2 ² ,1 ²)	9	-3	0	1	1	0	-1	0	1	-3	0
(2,1 ⁴)	5	-3	2	-1	1	0	0	1	-1	1	-1
(1 ⁶)	1	-1	1	-1	1	-1	1	-1	1	-1	1
\square_6	729	243	81	27	81	27	9	3	9	27	9

$$n(6) = 28$$

$$n(5,1) = 35$$

$$n(4,2) = 27$$

$$n(4,1^2) = 10$$

$$n(3^2) = 10$$

$$n(2^3) = 1$$

$$n(3,2,1) = 8$$

Π_7

CLASS	1^7	1^5_2	1^4_3	1^3_4	$1^3_2^2$	$1^2_2^2$	$1^2_3^2$	1^2_5	1_6	12_4	12^3	13^2	2_5	2^2_3	3_4	7
ORDER	1	21	70	210	105	420	504	840	630	105	280	504	210	420	720	
(7)	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
(6,1)	6	4	3	2	2	1	1	0	0	0	0	-1	-1	-1	-1	-1
(5,2)	14	6	2	0	2	0	-1	-1	0	2	-1	1	2	0	0	0
(5,1 ²)	15	5	3	1	-1	-1	0	0	-1	-3	0	0	-1	1	1	1
(4,3)	14	4	-1	-2	2	1	-1	0	0	0	2	-1	-1	1	0	0
(4,2,1)	35	5	-1	-1	-1	-1	0	1	1	1	-1	0	-1	-1	-1	0
(3 ² ,1)	21	1	-3	-1	1	1	1	0	-1	-3	0	1	1	-1	-1	0
(4,1 ³)	20	0	2	0	-4	0	0	0	0	0	2	0	2	0	0	-1
(3,2 ²)	21	-1	3	1	1	-1	1	0	-1	3	0	-1	1	1	1	0
(3,2,1 ²)	35	-5	1	1	-1	1	0	-1	1	-1	1	0	-1	1	1	0
(2 ³ ,1)	14	-4	-1	2	2	-1	-1	0	0	0	2	1	-1	-1	-1	0
(3,1 ⁴)	15	-5	3	-1	-1	1	0	0	-1	3	0	0	-1	-1	-1	1
(2 ² ,1 ³)	14	-6	2	0	2	0	-1	1	0	-2	-1	-1	2	0	0	0
(2,1 ⁵)	6	-4	3	-2	2	-1	1	0	0	0	0	1	-1	1	1	-1
(1 ⁷)	1	-1	1	-1	1	-1	1	-1	1	-1	1	-1	1	-1	1	1
$\overline{7}$	2187	729	243	81	243	81	27	9	27	81	27	9	27	9	3	3

$$n(7) = 36$$

$$n(4,3) = 24$$

$$n(6,1) = 48$$

$$n(4,2,1) = 15$$

$$n(5,2) = 42$$

$$n(3^2,1) = 6$$

$$n(5,1^2) = 15$$

$$n(3,2^2) = 3$$

Distribution of Irreducible Representations

Among the Eigenstates of S^2

N = 2

For $S = 2$, the irreducible representations are (2) ,

" $S = 1$, " " " " (1^2) ,

" $S = 0$, " " " " (2) .

N = 3

For $S = 3$, the irreducible representations are (3) ,

" $S = 2$, " " " " $(2,1)$,

" $S = 1$, " " " " $(2,1) + (3)$,

" $S = 0$, " " " " (1^3) .

N = 4

For $S = 4$, the irreducible representations are (4) ,

" $S = 3$, " " " " $(3,1)$,

" $S = 2$, " " " " $(3,1) + (2^2) + (4)$,

" $S = 1$, " " " " $(3,1) + (2,1^2)$,

" $S = 0$, " " " " $(2,1^2) + (4)$.

N = 5

For $S = 5$, the irreducible representations are (5) ,

" $S = 4$, " " " " $(4,1)$,

" $S = 3$, " " " " $(3,2) + (4,1) + (5)$,

" $S = 2$, " " " " $(3,1^2) + (3,2) + (4,1)$,

" $S = 1$, " " " " $(3,2) + (2^2,1) + (4,1) + (5)$,

" $S = 0$, " " " " $(3,1^2)$.

N = 6

For $S = 6$, the irreducible representations are (6) ,

"	$S = 5$,	"	"	"	"	$(5,1)$,
"	$S = 4$,	"	"	"	"	$(4,2) + (5,1) + (6)$,
"	$S = 3$,	"	"	"	"	$(4,1^2) + (4,2) + (5,1) + (3^2)$,
"	$S = 2$,	"	"	"	"	$(3,2,1) + 2(4,2) + (5,1) + (6)$,
"	$S = 1$,	"	"	"	"	$(3,2,1) + (4,1^2) + (5,1) + (3^2)$,
"	$S = 0$,	"	"	"	"	$(4,2) + (2^3) + (6)$.

N = 7





For $S = 7$, the irreducible representations are (7) ,

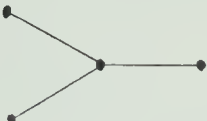

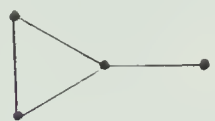

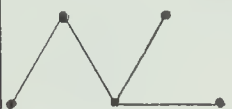


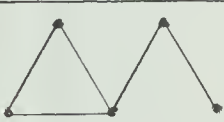



"	$S = 6$,	"	"	"	"	$(6,1)$,
"	$S = 5$,	"	"	"	"	$(5,2) + (6,1) + (7)$,
"	$S = 4$,	"	"	"	"	$(5,1^2) + (5,2) + (4,3) + (6,1)$,
"	$S = 3$,	"	"	"	"	$(4,2,1) + 2(5,2) + (4,3)$ $+ (6,1) + (7)$,
"	$S = 2$,	"	"	"	"	$(4,2,1) + (3^2,1) + (5,1^2)$ $+ (5,2) + (4,3) + (6,1)$,
"	$S = 1$,	"	"	"	"	$(4,2,1) + (3,2^2) + (5,2)$ $+ (4,3) + (6,1) + (7)$,
"	$S = 0$,	"	"	"	"	$(3^2,1) + (5,1^2)$.









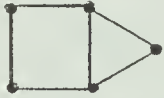


APPENDIX DCluster Expansion Data







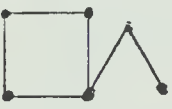




$$\psi_i, \phi_i \sim \sum_{n=0}^7 \frac{K^n}{3^n n!} a_n$$

- Note 1) Where two series are given for any cluster C_i , the upper series is ψ_i and the lower series is ϕ_i . If only one series is given, then it is ψ_i .
- 2) Only non-zero a_n are given in the table.
- 3) For the single point cluster C_0 , we have $\psi_0 = 2/3$ and $\phi_0 = \log 3$. For the single bond cluster C_1 , $a_1 = 4/3$ in ψ_1 .









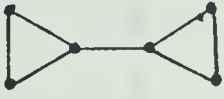













LABEL	GRAPH	a_2	a_3	a_4	a_5	a_6	a_7
C_1		-8/3	-16	+160	+448	-18,816	+40,704
		+8	-16	-96	+960	+2,688	-112,896
C_2		+8/3	-16	-24	+3,440	-38,904	-702,576
				-144	+1,440	+19,440	-514,080
C_3			+24	-1488	+1,080	+513,360	-2,005,416
			+48	+288	-13,680	+10,080	+3,922,128
C_4			+8	-48	-720	+17,040	-266,112
						+7,776	-181,440

LABEL	GRAPH	a_4	a_5	a_6	a_7
c_5		-144	+2,880	+28,080	-2,298,240
				+19,440	-408,240
c_6		+288	-8,960	-106,080	+6,014,400
		+192	+1,920	-85,824	-443,520
c_7		+64	-5,760	+136,080	+2,118,480
			-1,440	-11,232	+1,859,760
c_8		+32	-160	-9,192	+172,032
c_9			-480	+10,512	+149,688
c_{10}				+38,880	-1,360,800
c_{11}			-4,880	-216,960	+10,357,200
			+2,400	-84,384	-2,338,560
c_{12}			+320	-29,856	+271,488
					+108,864
c_{13}			+400	-22,992	+784,392
					+90,720
c_{14}			-480	-6,624	+2,351,664
					+272,160
c_{15}			+2,800	-66,960	-1,886,640
			+960	14,400	-772,120

LABEL	GRAPH	a_4	a_5	a_6	a_7
c_{16}			+720	-42,288 -8,640	+650,496 -90,720
c_{17}			+160	-480	-96,768
c_{18}				-2,880	+45,360
c_{19}				-1,440	+51,408
c_{20}					+136,080
c_{21}					+60,480
c_{22}				-77,760 -6,912	+645,120 -1,632,960
c_{23}				-27,792 +14,400	-1,448,496 -161,200
c_{24}				-21,936 +14,400	-2,323,104 -467,712
c_{25}				-19,488 -17,280	+1,485,120 -556,416
c_{26}				-23,136	-250,152 -226,800

LABEL	GRAPH	a_6	a_7
c_{27}		-6,144	-234,528 -169,344
c_{28}		+6,720	-397,824 -60,480
c_{29}		+27,360 +5,760	-491,904 +120,960
c_{30}		+2,400	-141,120
c_{31}		+2,400	-184,800
c_{32}		-2,880	-126,000
c_{33}		+4,320	-209,832
c_{34}		+2,400	-141,960
c_{35}			-4,032
c_{36}		-2,880	-41,328
c_{37}		+1,920	-202,272

LABEL	GRAPH	a_6	a_7
c_{38}			+272,160
c_{39}			-45,360
c_{40}			-84,672
c_{41}		+960	0
c_{42}			0
c_{43}			-20,160
c_{44}			-10,080
c_{45}		-----	-357,504 - 66,528
c_{46}		-----	-2,135,952 -665,280
c_{47}		-----	-955,416 +73,584
c_{48}			-272,160

LABEL	GRAPH	a_7	LABEL	GRAPH	a_7
c_{49}		$-113,232$ $+100,800$	c_{60}		$+12,264$
c_{50}		$-172,368$ $+100,800$	c_{61}		$-75,600$
c_{51}		$-242,928$ $-120,960$	c_{62}		$-110,880$
c_{52}		$-76,776$ $+100,800$	c_{63}		$-11,088$
c_{53}		$+26,880$	c_{64}		$-43,008$
c_{54}		$-37,968$	c_{65}		$-161,952$
c_{55}		$-120,960$	c_{66}		$-38,976$
c_{56}		$-140,616$	c_{67}		$-136,080$
c_{57}		$+15,624$	c_{68}		$+282,240$ $+40,320$
c_{58}		$-57,120$	c_{69}		$+63,840$
c_{59}		$-56,448$	c_{70}		$+47,040$

LABEL	GRAPH	a_7	LABEL	GRAPH	a_7
c_{71}		+30,240	c_{79}		-20,160
c_{72}		0	c_{80}		-20,160
c_{73}		+13,440	c_{81}		+16,800
c_{74}		-20,160	c_{82}		+16,800
c_{75}		+16,800	c_{83}		+16,800
c_{76}		0	c_{84}		-20,160
c_{77}		+16,800	c_{85}		+16,800
c_{78}		0	c_{86}		+6,720

Linear Combinations of ψ 's

$$\bar{\chi}_0(c_1, K) = 2\psi_0 + \psi_1$$

$$\bar{\chi}_0(c_2, K) = 3\psi_0 + 2\psi_1 + \psi_2$$

$$\bar{\chi}_0(c_3, K) = 3\psi_0 + 3\psi_1 + 3\psi_2 + \psi_3$$

$$\bar{\chi}_0(c_4, K) = 4\psi_0 + 3\psi_1 + 2\psi_2 + \psi_4$$

$$\overline{\chi}_0(c_5, K) = 4\psi_0 + 3\psi_1 + 3\psi_2 + \psi_5$$

$$\overline{\chi}_0(c_6, K) = 4\psi_0 + 4\psi_1 + 4\psi_2 + 4\psi_4 + \psi_6$$

$$\overline{\chi}_0(c_7, K) = 4\psi_0 + 4\psi_1 + 5\psi_2 + \psi_3 + 2\psi_4 + \psi_5 + \psi_7$$

$$\overline{\chi}_0(c_8, K) = 5\psi_0 + 4\psi_1 + 3\psi_2 + 2\psi_4 + \psi_8$$

$$\overline{\chi}_0(c_9, K) = 5\psi_0 + 4\psi_1 + 4\psi_2 + 2\psi_4 + \psi_5 + \psi_9$$

$$\overline{\chi}_0(c_{10}, K) = 5\psi_0 + 4\psi_1 + 6\psi_2 + 4\psi_5 + \psi_{10}$$

$$\overline{\chi}_0(c_{11}, K) = 4\psi_0 + 5\psi_1 + 8\psi_2 + 2\psi_3 + 6\psi_4 + 2\psi_5 + \psi_6 + 4\psi_7 + \psi_{11}$$

$$\overline{\chi}_0(c_{12}, K) = 5\psi_0 + 5\psi_1 + 6\psi_2 + \psi_3 + 4\psi_4 + \psi_5 + \psi_7 + 2\psi_8 + \psi_9 + \psi_{12}$$

$$\overline{\chi}_0(c_{13}, K) = 5\psi_0 + 5\psi_1 + 7\psi_2 + \psi_3 + 5\psi_4 + 2\psi_5 + 2\psi_7 + \psi_8 + 2\psi_9 + \psi_{13}$$

$$\overline{\chi}_0(c_{14}, K) = 5\psi_0 + 5\psi_1 + 8\psi_2 + \psi_3 + 4\psi_4 + 4\psi_5 + 2\psi_7 + 2\psi_9 + \psi_{10} + \psi_{14}$$

$$\overline{\chi}_0(c_{15}, K) = 5\psi_0 + 5\psi_1 + 5\psi_2 + 5\psi_4 + 5\psi_8 + \psi_{15}$$

$$\overline{\chi}_0(c_{16}, K) = 5\psi_0 + 5\psi_1 + 6\psi_2 + 6\psi_4 + \psi_5 + \psi_6 + 2\psi_8 + 2\psi_9 + \psi_{16}$$

$$\overline{\chi}_0(c_{17}, K) = 6\psi_0 + 5\psi_1 + 4\psi_2 + 3\psi_4 + 2\psi_8 + \psi_{17}$$

$$\overline{\chi}_0(c_{18}, K) = 6\psi_0 + 5\psi_1 + 5\psi_2 + 3\psi_4 + \psi_5 + 2\psi_8 + \psi_9 + \psi_{18}$$

$$\overline{\chi}_0(c_{19}, K) = 6\psi_0 + 5\psi_1 + 5\psi_2 + 4\psi_4 + \psi_5 + \psi_8 + 2\psi_9 + \psi_{19}$$

$$\overline{\chi}_0(c_{20}, K) = 6\psi_0 + 5\psi_1 + 7\psi_2 + 3\psi_4 + 4\psi_5 + 3\psi_9 + \psi_{10} + \psi_{20}$$

$$\overline{\chi}_0(c_{21}, K) = 6\psi_0 + 5\psi_1 + 6\psi_2 + 4\psi_4 + 2\psi_5 + 4\psi_9 + \psi_{21}$$

$$\begin{aligned}\overline{\chi}_0(c_{22}, K) &= 4\psi_0 + 6\psi_1 + 12\psi_2 + 4\psi_3 + 12\psi_4 + 4\psi_5 + 3\psi_6 + 12\psi_7 + \psi_8 \\ &\quad + 6\psi_{11} + \psi_{22}\end{aligned}$$

$$\overline{\chi}_0(c_{23}, K) = 5\psi_0 + 6\psi_1 + 9\psi_2 + 12\psi_4 + 2\psi_5 + 3\psi_6 + 6\psi_8 + 6\psi_9 + 6\psi_{16} + \psi_{23}$$

$$\begin{aligned}\overline{\chi}_0(c_{24}, K) &= 5\psi_0 + 6\psi_1 + 9\psi_2 + \psi_3 + 10\psi_4 + 2\psi_5 + \psi_6 + 2\psi_7 + 7\psi_8 + 4\psi_9 \\ &\quad + 2\psi_{12} + \psi_{13} + \psi_{15} + 2\psi_{16} + \psi_{24}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{25}, K) &= 5\psi_0 + 6\psi_1 + 10\psi_2 + 2\psi_3 + 8\psi_4 + 4\psi_5 + 4\psi_7 + 4\psi_8 + 4\psi_9 + \psi_{10} \\ &\quad + 4\psi_{12} + 2\psi_{14} + \psi_{25}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{26}, K) &= 5\psi_0 + 6\psi_1 + 11\psi_2 + 2\psi_3 + 10\psi_4 + 5\psi_5 + \psi_6 + 6\psi_7 + 2\psi_8 + 5\psi_9 \\ &\quad + \psi_{10} + \psi_{11} + 2\psi_{13} + 2\psi_{14} + \psi_{16} + \psi_{20}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{27}, K) &= 5\psi_0 + 6\psi_1 + 10\psi_2 + 2\psi_3 + 10\psi_4 + 3\psi_5 + \psi_6 + 5\psi_7 + 4\psi_8 + 4\psi_9 \\ &\quad + \psi_{11} + 2\psi_{12} + 2\psi_{13} + \psi_{16} + \psi_{27}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{28}, K) &= 6\psi_0 + 6\psi_1 + 7\psi_2 + 7\psi_4 + \psi_5 + 7\psi_8 + 2\psi_9 + \psi_{15} + 2\psi_{17} + 2\psi_{18} \\ &\quad + \psi_{19} + \psi_{28}\end{aligned}$$

$$\overline{\chi}_0(c_{29}, K) = 6\psi_0 + 6\psi_1 + 6\psi_2 + 6\psi_4 + 6\psi_8 + 6\psi_{17} + \psi_{29}$$

$$\overline{\chi}_0(c_{30}, K) = 6\psi_0 + 6\psi_1 + 8\psi_2 + 8\psi_4 + 2\psi_5 + \psi_6 + 6\psi_8 + 4\psi_9 + 2\psi_{16} + 4\psi_{18}$$

$$+\psi_{30}$$

$$\begin{aligned}\overline{\chi}_0(c_{31},K) &= 6\psi_0 + 6\psi_1 + 8\psi_2 + 9\psi_4 + 2\psi_5 + \psi_6 + 4\psi_8 + 6\psi_9 + 2\psi_{16} + \psi_{17} \\ &\quad + 2\psi_{19} + \psi_{21} + \psi_{31}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{32},K) &= 6\psi_0 + 6\psi_1 + 9\psi_2 + 8\psi_4 + 4\psi_5 + \psi_6 + 4\psi_8 + 6\psi_9 + \psi_{10} + 2\psi_{16} \\ &\quad + 2\psi_{18} + 2\psi_{20} + \psi_{32}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{33},K) &= 6\psi_0 + 6\psi_1 + 7\psi_2 + 8\psi_4 + \psi_5 + \psi_6 + 4\psi_8 + 3\psi_9 + \psi_{16} + 2\psi_{17} \\ &\quad + 2\psi_{19} + \psi_{33}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{34},K) &= 6\psi_0 + 6\psi_1 + 8\psi_2 + \psi_3 + 7\psi_4 + 2\psi_5 + 2\psi_7 + 4\psi_8 + 3\psi_9 + \psi_{12} \\ &\quad + \psi_{13} + \psi_{17} + \psi_{18} + \psi_{19} + \psi_{34}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{35},K) &= 6\psi_0 + 6\psi_1 + 8\psi_2 + \psi_3 + 6\psi_4 + 2\psi_5 + \psi_7 + 4\psi_8 + 4\psi_9 + 2\psi_{12} \\ &\quad + 2\psi_{18} + \psi_{21} + \psi_{35}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{36},K) &= 6\psi_0 + 6\psi_1 + 9\psi_2 + \psi_3 + 7\psi_4 + 4\psi_5 + 2\psi_7 + 2\psi_8 + 5\psi_9 + \psi_{10} \\ &\quad + \psi_{12} + \psi_{14} + 2\psi_{19} + \psi_{20} + \psi_{36}.\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{37},K) &= 6\psi_0 + 6\psi_1 + 7\psi_2 + \psi_3 + 5\psi_4 + \psi_5 + \psi_7 + 4\psi_8 + \psi_9 + \psi_{12} \\ &\quad + 2\psi_{17} + \psi_{18} + \psi_{37}\end{aligned}$$

$$\overline{\chi}_0(c_{38},K) = 6\psi_0 + 6\psi_1 + 12\psi_2 + \psi_3 + 6\psi_4 + 10\psi_5 + 3\psi_7 + 6\psi_9 + 5\psi_{10}$$

$$+ 3\psi_{14} + 2\psi_{20} + \psi_{38}$$

$$\begin{aligned}\overline{\chi}_0(c_{39}, K) &= 6\psi_0 + 6\psi_1 + 10\psi_2 + \psi_3 + 8\psi_4 + 5\psi_5 + 3\psi_7 + 2\psi_8 + 7\psi_9 + \psi_{10} \\ &\quad + 2\psi_{13} + \psi_{14} + \psi_{18} + \psi_{20} + \psi_{21} + \psi_{39}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{40}, K) &= 6\psi_0 + 6\psi_1 + 9\psi_2 + \psi_3 + 9\psi_4 + 3\psi_5 + 3\psi_7 + 3\psi_8 + 6\psi_9 + 3\psi_{13} \\ &\quad + 3\psi_{19} + \psi_{40}\end{aligned}$$

$$\overline{\chi}_0(c_{41}, K) = 7\psi_0 + 6\psi_1 + 5\psi_2 + 4\psi_4 + 3\psi_8 + 2\psi_{17} + \psi_{41}$$

$$\overline{\chi}_0(c_{42}, K) = 7\psi_0 + 6\psi_1 + 6\psi_2 + 6\psi_4 + \psi_5 + 3\psi_8 + 3\psi_9 + 3\psi_{19} + \psi_{42}$$

$$\overline{\chi}_0(c_{43}, K) = 7\psi_0 + 6\psi_1 + 6\psi_2 + 4\psi_4 + \psi_5 + 3\psi_8 + \psi_9 + 2\psi_{17} + \psi_{18} + \psi_{43}$$

$$\overline{\chi}_0(c_{44}, K) = 7\psi_0 + 6\psi_1 + 6\psi_2 + 5\psi_4 + \psi_5 + 3\psi_8 + 2\psi_9 + \psi_{17} + \psi_{18} + \psi_{19} + \psi_{44}$$

$$\begin{aligned}\overline{\chi}_0(c_{45}, K) &= 5\psi_0 + 7\psi_1 + 13\psi_2 + 2\psi_3 + 18\psi_4 + 4\psi_5 + 3\psi_6 + 6\psi_7 + 14\psi_8 \\ &\quad + \psi_{11} + 10\psi_9 + 6\psi_{12} + 4\psi_{13} + 2\psi_{15} + 8\psi_{16} + \psi_{23} + 4\psi_{24} \\ &\quad + 2\psi_{27} + \psi_{45}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{46}, K) &= 5\psi_0 + 7\psi_1 + 15\psi_2 + 3\psi_3 + 18\psi_4 + 8\psi_5 + 3\psi_6 + 12\psi_7 + 6\psi_8 \\ &\quad + 12\psi_9 + 2\psi_{10} + 3\psi_{11} + 6\psi_{13} + 6\psi_{14} + 6\psi_{16} + \psi_{23} + 6\psi_{26} \\ &\quad + \psi_{46}\end{aligned}$$

$$\overline{\chi}_0(c_{47}, K) = 5\psi_0 + 7\psi_1 + 14\psi_2 + 3\psi_3 + 17\psi_4 + 6\psi_5 + 2\psi_6 + 10\psi_7 + 10\psi_8$$

$$\begin{aligned}
& + 10\psi_9 + \psi_{10} + 2\psi_{11} + 6\psi_{12} + 5\psi_{13} + 3\psi_{14} + \psi_{15} + 4\psi_{16} \\
& + 2\psi_{24} + \psi_{25} + 2\psi_{26} + 2\psi_{27} + \psi_{47}
\end{aligned}$$

$$\begin{aligned}
\overline{\chi}_0(c_{48}, K) &= 5\psi_0 + 7\psi_1 + 15\psi_2 + 4\psi_3 + 18\psi_4 + 7\psi_5 + 3\psi_6 + 15\psi_7 + 6\psi_8 \\
&+ 9\psi_9 + \psi_{10} + 6\psi_{11} + 3\psi_{12} + 6\psi_{13} + 3\psi_{14} + 3\psi_{16} + \psi_{22} \\
&+ 3\psi_{26} + 3\psi_{27} + \psi_{48}
\end{aligned}$$

$$\begin{aligned}
\overline{\chi}_0(c_{49}, K) &= 6\psi_0 + 7\psi_1 + 10\psi_2 + 14\psi_4 + 2\psi_5 + 2\psi_6 + 10\psi_8 + 8\psi_9 + 4\psi_{16} \\
&+ 8\psi_{17} + 6\psi_{19} + \psi_{21} + \psi_{29} + 2\psi_{31} + 4\psi_{33} + \psi_{49}
\end{aligned}$$

$$\begin{aligned}
\overline{\chi}_0(c_{50}, K) &= 6\psi_0 + 7\psi_1 + 10\psi_2 + 13\psi_4 + 2\psi_5 + \psi_6 + 14\psi_8 + 6\psi_9 + 2\psi_{15} \\
&+ 2\psi_{16} + 6\psi_{17} + 6\psi_{18} + 4\psi_{19} + 4\psi_{28} + \psi_{30} + 2\psi_{33} + \psi_{50}
\end{aligned}$$

$$\begin{aligned}
\overline{\chi}_0(c_{51}, K) &= 6\psi_0 + 7\psi_1 + 11\psi_2 + \psi_3 + 12\psi_4 + 4\psi_5 + \psi_6 + 2\psi_7 + 8\psi_8 \\
&+ 8\psi_9 + \psi_{10} + 2\psi_{12} + \psi_{14} + 2\psi_{16} + 4\psi_{17} + 2\psi_{18} + 4\psi_{19} \\
&+ 2\psi_{20} + \psi_{32} + 2\psi_{33} + 2\psi_{36} + 2\psi_{37} + \psi_{51}
\end{aligned}$$

$$\begin{aligned}
\overline{\chi}_0(c_{52}, K) &= 6\psi_0 + 7\psi_1 + 10\psi_2 + \psi_3 + 11\psi_4 + 2\psi_5 + 2\psi_7 + 12\psi_8 + 4\psi_9 \\
&+ 2\psi_{12} + \psi_{13} + \psi_{15} + 8\psi_{17} + 4\psi_{18} + 2\psi_{19} + 2\psi_{28} + 2\psi_{34} \\
&+ 2\psi_{37} + \psi_{52}
\end{aligned}$$

$$\overline{\chi}_0(c_{53}, K) = 6\psi_0 + 7\psi_1 + 10\psi_2 + 2\psi_3 + 8\psi_4 + 2\psi_5 + 2\psi_7 + 8\psi_8 + 4\psi_9$$

$$+ 4\psi_{12} + 4\psi_{17} + 4\psi_{18} + \psi_{22} + 2\psi_{35} + 4\psi_{37} + \psi_{53}$$

$$\begin{aligned}\overline{\chi}_0(c_{54}, K) = & 6\psi_0 + 7\psi_1 + 12\psi_2 + 2\psi_3 + 12\psi_4 + 5\psi_5 + 5\psi_7 + 8\psi_8 + 9\psi_9 \\ & + \psi_{10} + 5\psi_{12} + 2\psi_{13} + 2\psi_{14} + 2\psi_{17} + 3\psi_{18} + 2\psi_{19} + \psi_{20} \\ & + \psi_{21} + \psi_{25} + 2\psi_{34} + \psi_{35} + \psi_{36} + \psi_{37} + \psi_{39} + \psi_{54}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{55}, K) = & 6\psi_0 + 7\psi_1 + 14\psi_2 + 2\psi_3 + 12\psi_4 + 10\psi_5 + 6\psi_7 + 4\psi_8 + 12\psi_9 \\ & + 5\psi_{10} + 4\psi_{12} + 6\psi_{14} + 4\psi_{19} + 4\psi_{20} + \psi_{25} + 4\psi_{36} + 2\psi_{38} \\ & + \psi_{55}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{56}, K) = & 6\psi_0 + 7\psi_1 + 12\psi_2 + \psi_3 + 14\psi_4 + 5\psi_5 + \psi_6 + 3\psi_7 + 10\psi_8 \\ & + 11\psi_9 + \psi_{10} + 2\psi_{12} + 2\psi_{13} + \psi_{14} + \psi_{15} + 3\psi_{16} + 2\psi_{17} \\ & + 3\psi_{18} + 3\psi_{19} + 2\psi_{20} + \psi_{21} + \psi_{24} + \psi_{28} + \psi_{31} + \psi_{32} + \psi_{34} \\ & + \psi_{36} + \psi_{39} + \psi_{56}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{57}, K) = & 6\psi_0 + 7\psi_1 + 11\psi_2 + \psi_3 + 13\psi_4 + 3\psi_5 + \psi_6 + 2\psi_7 + 12\psi_8 \\ & + 8\psi_9 + 3\psi_{12} + \psi_{13} + \psi_{15} + 3\psi_{16} + 3\psi_{17} + 5\psi_{18} + 2\psi_{19} \\ & + \psi_{21} + \psi_{24} + \psi_{28} + \psi_{30} + \psi_{31} + \psi_{34} + \psi_{35} + \psi_{37} + \psi_{57}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{58}, K) = & 6\psi_0 + 7\psi_1 + 11\psi_2 + \psi_3 + 14\psi_4 + 3\psi_5 + \psi_6 + 3\psi_7 + 11\psi_8 \\ & + 8\psi_9 + 2\psi_{12} + 3\psi_{13} + \psi_{15} + 2\psi_{16} + 4\psi_{17} + 2\psi_{18} + 5\psi_{19}\end{aligned}$$

$$+ \psi_{24} + \psi_{28} + 2\psi_{33} + 2\psi_{34} + \psi_{40} + \psi_{58}$$

$$\begin{aligned} \overline{\chi}_0(c_{59}, K) = & 6\psi_0 + 7\psi_1 + 13\psi_2 + 2\psi_3 + 14\psi_4 + 6\psi_5 + \psi_6 + 6\psi_7 + 8\psi_8 + 12\psi_9 \\ & + \psi_{10} + \psi_{11} + 4\psi_{12} + 4\psi_{13} + \psi_{14} + 2\psi_{16} + 4\psi_{18} + 2\psi_{20} + 2\psi_{21} \\ & + 2\psi_{27} + \psi_{32} + 2\psi_{35} + 2\psi_{39} + \psi_{59} \end{aligned}$$

$$\begin{aligned} \overline{\chi}_0(c_{60}, K) = & 6\psi_0 + 7\psi_1 + 13\psi_2 + 3\psi_3 + 15\psi_4 + 6\psi_5 + \psi_6 + 7\psi_7 + 7\psi_8 + 12\psi_9 \\ & + \psi_{10} + \psi_{11} + 2\psi_{12} + 5\psi_{13} + 2\psi_{14} + 2\psi_{16} + \psi_{17} + \psi_{18} + 4\psi_{19} \\ & + \psi_{20} + \psi_{21} + \psi_{26} + \psi_{27} + \psi_{31} + \psi_{34} + \psi_{36} + \psi_{39} + \psi_{40} + \psi_{60} \end{aligned}$$

$$\begin{aligned} \overline{\chi}_0(c_{61}, K) = & 6\psi_0 + 7\psi_1 + 15\psi_2 + 2\psi_3 + 14\psi_4 + 11\psi_5 + \psi_6 + 8\psi_7 + 4\psi_8 + 14\psi_9 \\ & + 5\psi_{10} + \psi_{11} + 4\psi_{13} + 6\psi_{14} + 2\psi_{16} + 2\psi_{18} + 4\psi_{20} + \psi_{21} \\ & + 2\psi_{26} + \psi_{32} + 2\psi_{38} + 2\psi_{39} + \psi_{61} \end{aligned}$$

$$\begin{aligned} \overline{\chi}_0(c_{62}, K) = & 6\psi_0 + 7\psi_1 + 14\psi_2 + 2\psi_3 + 15\psi_4 + 8\psi_5 + \psi_6 + 8\psi_7 + 6\psi_8 + 14\psi_9 \\ & + 2\psi_{10} + \psi_{11} + 6\psi_{13} + 4\psi_{14} + 2\psi_{16} + 4\psi_{18} + 2\psi_{20} + 2\psi_{21} + 2\psi_{26} \\ & + 2\psi_{30} + 4\psi_{39} + \psi_{62} \end{aligned}$$

$$\begin{aligned} \overline{\chi}_0(c_{63}, K) = & 6\psi_0 + 7\psi_1 + 12\psi_2 + 2\psi_3 + 14\psi_4 + 4\psi_5 + \psi_6 + 6\psi_7 + 10\psi_8 \\ & + 8\psi_9 + \psi_{11} + 4\psi_{12} + 4\psi_{13} + 2\psi_{16} + 2\psi_{17} + 4\psi_{18} + 2\psi_{19} \\ & + 2\psi_{27} + \psi_{30} + 4\psi_{34} + \psi_{63} \end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{64}, K) = & 6\psi_0 + 7\psi_1 + 11\psi_2 + 2\psi_3 + 12\psi_4 + 3\psi_5 + \psi_6 + 5\psi_7 + 8\psi_8 + 5\psi_9 \\ & + \psi_{11} + 3\psi_{12} + 2\psi_{13} + \psi_{16} + 4\psi_{17} + 2\psi_{18} + 2\psi_{19} + \psi_{27} + \psi_{33} \\ & + 2\psi_{34} + 2\psi_{37} + \psi_{64}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{65}, K) = & 6\psi_0 + 7\psi_1 + 12\psi_2 + 2\psi_3 + 13\psi_4 + 5\psi_5 + \psi_6 + 6\psi_7 + 6\psi_8 + 8\psi_9 \\ & + \psi_{10} + \psi_{11} + 2\psi_{12} + 2\psi_{13} + 2\psi_{14} + \psi_{16} + 2\psi_{17} + \psi_{18} + 4\psi_{19} \\ & + \psi_{20} + \psi_{26} + \psi_{33} + 2\psi_{34} + 2\psi_{36} + \psi_{65}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{66}, K) = & 6\psi_0 + 7\psi_1 + 11\psi_2 + 16\psi_4 + 3\psi_5 + 3\psi_6 + 10\psi_8 + 12\psi_9 + \\ & + 8\psi_{16} + 4\psi_{17} + 6\psi_{19} + 2\psi_{21} + \psi_{23} + 4\psi_{31} + 2\psi_{33} + \psi_{66}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{67}, K) = & 6\psi_0 + 7\psi_1 + 12\psi_2 + 15\psi_4 + 5\psi_5 + 3\psi_6 + 12\psi_8 + 12\psi_9 + \psi_{10} \\ & + 9\psi_{16} + 9\psi_{18} + 3\psi_{20} + \psi_{23} + 3\psi_{30} + 3\psi_{32} + \psi_{67}\end{aligned}$$

$$\overline{\chi}_0(c_{68}, K) = 7\psi_0 + 7\psi_1 + 7\psi_2 + 7\psi_4 + 7\psi_8 + 7\psi_{17} + 7\psi_{41} + \psi_{68}$$

$$\begin{aligned}\overline{\chi}_0(c_{69}, K) = & 7\psi_0 + 7\psi_1 + 8\psi_2 + 8\psi_4 + \psi_5 + 8\psi_8 + 2\psi_9 + 8\psi_{17} + 2\psi_{18} + \psi_{19} \\ & + \psi_{29} + 2\psi_{31} + 2\psi_{33} + 2\psi_{34} + \psi_{69}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{70}, K) = & 7\psi_0 + 7\psi_1 + 8\psi_2 + 9\psi_4 + \psi_5 + 9\psi_8 + 3\psi_9 + \psi_{15} + 4\psi_{17} + 2\psi_{18} \\ & + 3\psi_{19} + \psi_{28} + 2\psi_{31} + \psi_{32} + 2\psi_{33} + \psi_{70}\end{aligned}$$

$$\overline{\chi}_0(c_{71}, K) = 7\psi_0 + 7\psi_1 + 8\psi_2 + 9\psi_4 + \psi_5 + \psi_6 + 6\psi_8 + 3\psi_9 + \psi_{16} + 4\psi_{17}$$

$$+ \psi_{18} + 2\psi_{19} + \psi_{33} + 2\psi_{41} + 2\psi_{44} + \psi_{71}$$

$$\begin{aligned} \overline{\chi}_0(c_{72}, K) &= 7\psi_0 + 7\psi_1 + 9\psi_2 + 10\psi_4 + 2\psi_5 + \psi_6 + 6\psi_8 + 6\psi_9 + \psi_{16} + 4\psi_{17} \\ &\quad + 2\psi_{18} + 4\psi_{19} + \psi_{21} + 2\psi_{33} + 2\psi_{43} + \psi_{72} \end{aligned}$$

$$\begin{aligned} \overline{\chi}_0(c_{73}, K) &= 7\psi_0 + 7\psi_1 + 8\psi_2 + \psi_3 + 6\psi_4 + \psi_5 + \psi_7 + 5\psi_8 + \psi_9 + \psi_{12} \\ &\quad + 4\psi_{17} + \psi_{18} + \psi_{37} + 2\psi_{41} + \psi_{43} + \psi_{73} \end{aligned}$$

$$\begin{aligned} \overline{\chi}_0(c_{74}, K) &= 7\psi_0 + 7\psi_1 + 10\psi_2 + 11\psi_4 + 4\psi_5 + \psi_6 + 6\psi_8 + 9\psi_9 + \psi_{10} \\ &\quad + 2\psi_{16} + 2\psi_{17} + 2\psi_{18} + 4\psi_{19} + 3\psi_{21} + \psi_{32} + \psi_{33} + 2\psi_{44} + \psi_{74} \end{aligned}$$

$$\begin{aligned} \overline{\chi}_0(c_{75}, K) &= 7\psi_0 + 7\psi_1 + 9\psi_2 + 11\psi_4 + 2\psi_5 + \psi_6 + 7\psi_8 + 7\psi_9 + 2\psi_{16} \\ &\quad + 3\psi_{17} + \psi_{18} + 5\psi_{19} + \psi_{21} + \psi_{31} + \psi_{33} + \psi_{41} + \psi_{42} + \psi_{44} \\ &\quad + \psi_{75} \end{aligned}$$

$$\begin{aligned} \overline{\chi}_0(c_{76}, K) &= 7\psi_0 + 7\psi_1 + 9\psi_2 + \psi_3 + 6\psi_4 + 2\psi_5 + \psi_7 + 6\psi_8 + 2\psi_9 + \psi_{12} \\ &\quad + 4\psi_{17} + 4\psi_{18} + 2\psi_{37} + 2\psi_{43} + \psi_{76} \end{aligned}$$

$$\begin{aligned} \overline{\chi}_0(c_{77}, K) &= 7\psi_0 + 7\psi_1 + 9\psi_2 + 10\psi_4 + 2\psi_5 + \psi_6 + 8\psi_8 + 5\psi_9 + 2\psi_{16} + 4\psi_{17} \\ &\quad + 4\psi_{18} + 2\psi_{19} + \psi_{30} + \psi_{33} + 2\psi_{43} + 2\psi_{44} + \psi_{77} \end{aligned}$$

$$\begin{aligned} \overline{\chi}_0(c_{78}, K) &= 7\psi_0 + 7\psi_1 + 9\psi_2 + \psi_3 + 8\psi_4 + 2\psi_5 + \psi_7 + 6\psi_8 + 5\psi_9 + 2\psi_{12} \\ &\quad + 2\psi_{17} + 3\psi_{18} + 2\psi_{19} + \psi_{21} + \psi_{35} + \psi_{37} + 2\psi_{44} + \psi_{78} \end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{79}, K) = & 7\psi_0 + 7\psi_1 + 10\psi_2 + \psi_3 + 8\psi_4 + 4\psi_5 + 2\psi_7 + 5\psi_8 + 5\psi_9 + \psi_{10} \\ & + \psi_{12} + \psi_{14} + 2\psi_{17} + 3\psi_{18} + 2\psi_{19} + \psi_{20} + \psi_{36} + 2\psi_{44} \\ & + \psi_{79}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{80}, K) = & 7\psi_0 + 7\psi_1 + 10\psi_2 + 9\psi_4 + 4\psi_5 + 9\psi_8 + 6\psi_9 + \psi_{10} + \psi_{15} + 4\psi_{17} \\ & + 6\psi_{18} + 2\psi_{19} + 2\psi_{20} + 2\psi_{28} + 2\psi_{43} + \psi_{80}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{81}, K) = & 7\psi_0 + 7\psi_1 + 9\psi_2 + \psi_3 + 8\psi_4 + 2\psi_5 + 2\psi_7 + 6\psi_8 + 3\psi_9 + \psi_{12} \\ & + \psi_{13} + 4\psi_{17} + 2\psi_{18} + \psi_{19} + \psi_{34} + \psi_{37} + \psi_{41} + \psi_{43} + \psi_{44} \\ & + \psi_{81}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{82}, K) = & 7\psi_0 + 7\psi_1 + 9\psi_2 + 10\psi_4 + 2\psi_5 + 9\psi_8 + 6\psi_9 + \psi_{15} + 4\psi_{17} \\ & + 4\psi_{18} + 4\psi_{19} + \psi_{21} + 2\psi_{28} + \psi_{41} + 2\psi_{44} + \psi_{82}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{83}, K) = & 7\psi_0 + 7\psi_1 + 9\psi_2 + 9\psi_4 + 2\psi_5 + 10\psi_8 + 4\psi_9 + \psi_{15} + 5\psi_{17} \\ & + 6\psi_{18} + 2\psi_{19} + 2\psi_{28} + 2\psi_{43} + 2\psi_{44} + \psi_{83}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{84}, K) = & 7\psi_0 + 7\psi_1 + 10\psi_2 + \psi_3 + 10\psi_4 + 4\psi_5 + 2\psi_7 + 5\psi_8 + 8\psi_9 + \psi_{10} \\ & + 2\psi_{12} + \psi_{14} + 6\psi_{19} + 2\psi_{20} + 2\psi_{36} + 2\psi_{42} + \psi_{84}\end{aligned}$$

$$\begin{aligned}\overline{\chi}_0(c_{85}, K) = & 7\psi_0 + 7\psi_1 + 9\psi_2 + \psi_3 + 9\psi_4 + 2\psi_5 + 2\psi_7 + 7\psi_8 + 4\psi_9 + 2\psi_{12} \\ & + \psi_{13} + 3\psi_{17} + 2\psi_{18} + 2\psi_{19} + 2\psi_{34} + \psi_{41} + 2\psi_{44} + \psi_{85}\end{aligned}$$

The series for $\log Z(C_i, K)$ in terms of ϕ_j $j < i$ are, of course, identical to the above series.

$$2(\text{Diagram 1}) = 10(\text{Diagram 2}) - 2(\text{Diagram 3}) - 2(\text{Diagram 4})$$

$$(\text{Diagram 5}) = 9(\text{Diagram 2}) - 2(\text{Diagram 6}) - (\text{Diagram 7})$$

$$(\text{Diagram 8}) = 20(\text{Diagram 2}) - 2(\text{Diagram 3}) - 4(\text{Diagram 9})$$

$$- 2(\text{Diagram 10})$$

$$(\text{Diagram 11}) = 10(\text{Diagram 2}) - (\text{Diagram 7}) - 2(\text{Diagram 4})$$

$$- 2(\text{Diagram 12})$$

$$(\text{Diagram 13}) = 11(\text{Diagram 14}) - 4(\text{Diagram 15}) - 2(\text{Diagram 9})$$

$$- 2(\text{Diagram 16})$$

$$2(\text{Diagram 17}) = 10(\text{Diagram 14}) - (\text{Diagram 15}) - (\text{Diagram 18})$$

$$(\text{Diagram 19}) = 10(\text{Diagram 14}) - 4(\text{Diagram 15}) - 2(\text{Diagram 18})$$

$$(\text{Diagram 20}) = 9(\text{Diagram 14}) - (\text{Diagram 15}) - 2(\text{Diagram 9})$$

$$(\text{Diagram 21}) = 20(\text{Diagram 14}) - 2(\text{Diagram 18}) - (\text{Diagram 19})$$

$$- 2(\text{Diagram 16})$$

$$2(\text{diagram}) = 11(\text{diagram}) - (\text{diagram}) - 2(\text{diagram})$$

$$- 2(\text{diagram})$$

$$2(\text{diagram}) = 11(\text{diagram}) - 2(\text{diagram}) - 2(\text{diagram})$$

$$- (\text{diagram}) - 2(\text{diagram})$$

$$2(\text{diagram}) = 22(\text{diagram}) - 2(\text{diagram}) - 2(\text{diagram})$$

$$- 2(\text{diagram}) - 2(\text{diagram}) - 14(\text{diagram})$$

Numerical Values of Lattice Constants

$$(\text{diagram}) = 7,776$$

$$(\text{diagram}) = 3,744$$

$$(\text{diagram}) = 1,632$$

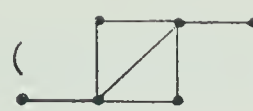



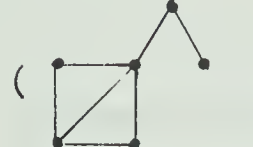




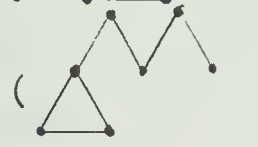





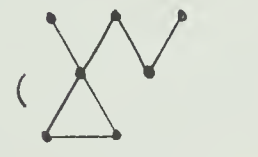
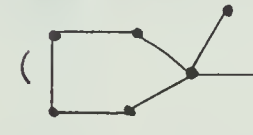
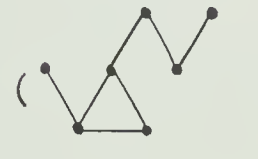
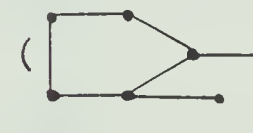





$$(\text{diagram}) = 2,592$$

$$(\text{diagram}) = 7,392$$

$$(\text{diagram}) = 3,024$$

$$(\text{diagram}) = 6,720$$

$$(\text{diagram}) = 3,324$$

	$= 2,844$		$= 58,992$
	$= 12,144$		$= 104,016$
	$= 6,600$		$= 229,176$
	$= 7,176$		$= 117,672$
	$= 984$		$= 266,928$
	$= 600$		$= 121,776$
	$= 51,468$		$= 235,584$
	$= 76,752$		$= 220,992$
	$= 30,984$		$= 490,896$
	$= 66,480$		$= 106,536$
	$= 68,064$		$= 242,808$
	$= 131,040$		$= 8,086,074$

APPENDIX F

The Triangular Lattice

In the notation of appendix D, the zero field susceptibility series is given by,

$$a_0 = +2/3$$

$$a_4 = +3240$$

$$a_1 = +4$$

$$a_5 = +31,584$$

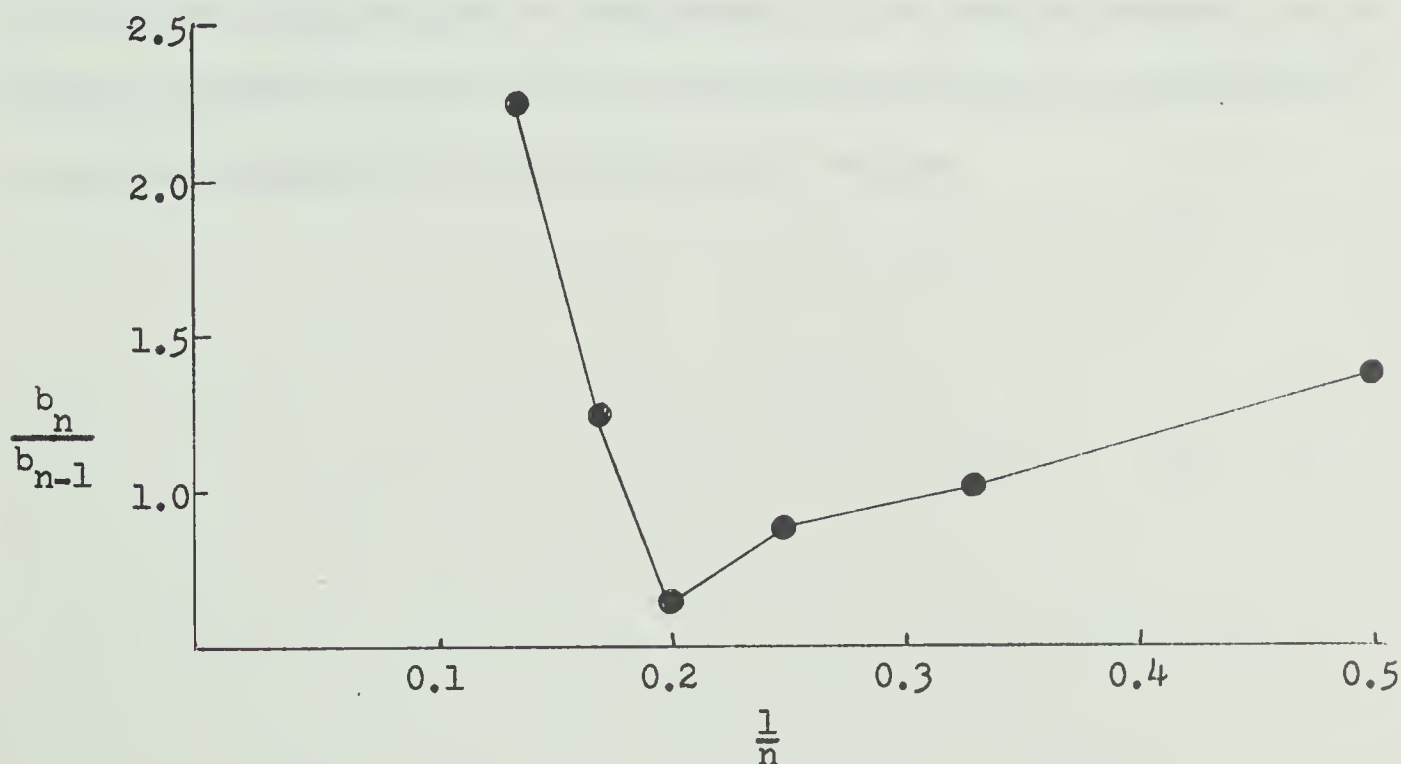
$$a_2 = +32$$

$$a_6 = +724,032$$

$$a_3 = +312$$

$$a_7 = +34,031,232$$

The Domb ratio plot gives,



and the Padé approximant table for K_c from $\frac{d}{dK} \log \chi_0(K)$ is of the form,

$\begin{array}{c} M \\ \backslash \\ N \end{array}$	0	1	2	3	4	5
1					N.P.R.	0.60
2			N.R.R.	N.R.R.	N.R.R.	
3			1.59	2.24		
4		N.P.R.	N.R.R.			
5	0.70	0.68				
6						

where N.R.P. = no real root and N.P.R. = no positive real root.

The manner in which the above ratio plot contrasts with Figure 6.1 and the complete lack of consistency of the Padé approximant table does not permit us to draw any conclusions regarding the existence of a critical temperature in the triangular lattice.

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